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TRITIUM AND NOBLE-GAS FISSION PRODUCTS IN THE NUCLEAR FUEL CYCLE. I. REACTORS

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF TABLES (cond.)

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ABSTRACT

A review of the behavior of tritium and noble-gas fission products in nuclear reactors is presented. The sources of tritium considered include fission and activation of poisons and impurities in coolants. The noble gases included in this review are limited to fission products, with emphasis on the long-lived species. Reactor types surveyed include light water reactors, high-temperature gas-cooled reactors, and liquid-metal-cooled fast breeder reactors. Data indicative of the normal operating procedures have been expanded to provide estimates of the quantities of tritium and noble gases and their diluents expected at various points along the flow path for both gaseous and liquid waste streams. Data are normalized to an energy of 1000 MWe-yr to permit comparison with present-generation reactors.

I. INTRODUCTION

Tritium and noble-gas fission products represent a major part of the radioactive material released to the environment in the generation of power by nuclear fission. As the attention paid to environmental affairs increased, the need for information on the details of the behavior of tritium and noble gases became more pressing. Furthermore, systematic investigations of means to reduce or eliminate releases of radioactive materials to the environment required information on the source of such materials and an identification of volumes, flow rates, and compositions of streams in which radioactivity is carried.

The present report is the first in a series which summarizes information available in the literature dealing with the behavior of tritium and noble gases. A summary report recently issued (WASH-1258) is the source of some of the data presented herein. Field studies by the U. S. Atomic Energy Commission (AEC), the Environmental Protection Agency (EPA), and the utilities continue to produce data on the release of radioactivity; the data from the more recent of their reports have not been incorporated into this report.

The data presented herein are designed to provide a summary of the chemical environment of tritium and noble gases as generated in, transported in, and released from four major types of reactors: (1) boiling water reactors (BWRs), (2) pressurized water reactors (PWRs), (3) high-temperature gas-cooled reactors (HTGRs), and (4) liquid-metal-cooled fast breeder reactors (LMFBRs). Changes in design of fuel elements, off-gas systems, water-management schemes, etc., will effect, in individual cases, significant changes in the rates and amounts of radioactive materials released. In the

absence of such detailed information, the present report may provide the reader with a generalized description of the generation and subsequent disposition of tritium and noble gases in nuclear power plants. Pertinent physical and radiochemical data for tritium and noble gases are summarized in the Appendix.

The formation of noble gases (xenon and krypton) in nuclear reactors occurs by fission in nuclear fuel. The formation of tritium occurs by fission in nuclear fuel and also by neutron activation reactions, mainly with lithium and boron isotopes (Kouts; Locante 1971, 1973; Smith).

The appearance of noble gases in the effluent streams of a reactor results mainly from fuel-cladding failure. The appearance of tritium in effluent streams results from cladding failure and from neutron activation of lithium and boron dissolved in or in direct contact with primary coolant; moreover, if the cladding is stainless steel, diffusion of tritium through the cladding can occur.

The disposal of tritium and noble gases in reactor waste streams is usually handled in present operations by diluting them with air or water to reduce their concentrations to values below the concentration guidelines for public exposure (10 CFR 20) and discharging them to the environment; disposal in the near future is expected to be conducted similarly. Recent activities related to reducing environmental discharges have included development of improved reactor-management techniques, monitoring of reactor effluent streams both by reactor operators and by federal and local governmental agencies, and amendment of regulations on concentrations in effluents in light-water reactors (10 CFR 50).

Because of recent concern that dilution and dispersal to the environment may eventually lead to a buildup of the long-lived isotopes tritium (12.3 yr) and ^{85}Kr (10.8 yr) that may be biologically hazardous, present dispersal practices have been extensively discussed (Bond, Coleman, Hendrickson, Karol, Koranda, Nichols, Pigford, Whitton). Although no definitive conclusions have been reached, the possibility exists that future prohibition of dispersal to the environment could result from rational proof of the hazard or from opinion and political action only. This possibility has stimulated work on development of processes for isolation of noble gases from effluent streams and subsequent storage (Dunster, Kovach, Nichols, Slansky). Collection and storage processes are not discussed in this report. Although buildup of tritium has also been debated extensively (Koranda, Pigford), its isolation and collection seems to have received less attention than similar processes for ^{85}Kr.

The purpose of this report is to characterize the reactor effluent streams that would be encountered in collection or isolation processes. A brief description of each type of reactor, sufficient only to illustrate pathways available to tritium and noble gases, is presented. The effluent streams are described in terms of amounts of tritium and noble gases and the amounts of major accompanying constituents. Estimates of materials in effluents are expressed in amounts per 1000 MWe-yr to provide a common basis of comparison for the various reactor types. Amounts of fission products generated per 1000 MWe-yr were derived from known yields per fission (Dudey, Katcoff), an assumption of 2.7×10^{21} fissions per MWt-day (based on

200 MeV/fission), and an assumption that the thermal-to-electrical conversion efficiency is 33% for light-water reactors and about 40% for LMFBRs and HTGRs. Amounts of other effluent-stream constituents are based on estimated system volumes, waste volumes, and flow rates for reactors of the 1000-MWe size. Full characterization of effluent streams at reactors of the 1000-MWe size must await further operational experience and surveillance. Experience in full-power operation and detailed monitoring results (Kahn, Kahn 1971) available at present are mainly based on smaller, first-generation light-water reactors.

The types of reactors discussed here are those that currently seem most likely to be important in civilian power generation. Light-water thermal reactors (BWRs and PWRs) have been included because planning and construction activities indicate that they will produce a large amount of electrical power by 1982, including 49,000 Mwe for BWRs and 88,000 MWe for PWRs (TID-8200-R27). The HTGR has been included because the prospects of its making a significant contribution to U. S. generating capacity seem good on the basis of construction of one 330-MWe plant and orders for the purchase of two 1140-NWe plants and four 770-MWe plants (TID-8200-R27). The LMFBR has been included on the basis of the current intensive effort in the U. S. to develop this type of reactor for future civilian power generation.

The many short-lived gaseous isotopes (with half-lives ranging from a few seconds to 12 days) formed in reactors by fission and by activation of air are given little attention in this report. Although their appearance in reactor effluent streams is significant for reactor environs, these isotopes present no buildup problems and will probably be controlled most economically by delay-decay techniques. Thus, the problems emphasized here are related to the control of hazards associated with the longer-lived isotopes, 10.8-yr ^{85}Kr and 12.3-yr ^{3}H.

II. BEHAVIOR OF TRITIUM AND ^{85}Kr IN BOILING WATER REACTORS

The appearance of significant quantities of tritium and noble gases in effluent waste streams of a BWR begins with their introduction into the primary coolant, mainly by diffusion through defects in Zircaloy cladding of the fuel (Smith). The tritium is expected to be converted rapidly into THO by exchange with H_2O (Ray). Thus, water is the main vehicle for tritium in the BWR system, and tritium can be discharged from the plant in liquid waste streams or as water vapor in gaseous waste streams. The noble gases are expected to have low solubility in the boiling primary coolant and are expected to be discharged in gaseous waste streams. Descriptions of the coolant systems and the gaseous and liquid waste streams of the BWR are given below.

A. Boiling Water Reactor System Descriptions

1. Coolant Cycle

Figure 1 is a diagram of the primary coolant cycle of a typical BWR. Water is circulated through the core,* and the resulting steam-water mixture passes from the core through steam separators and dryers contained in the head of the reactor vessel, thereby reducing the moisture content of the steam to a maximum of 0.3%. The steam enters the high-pressure turbine casing at 1050 psi, flows through additional moisture-separator units, and into the low-pressure casings of the turbine. From the turbines, the steam flows into the main condenser. Condensate from the main condenser passes through filters, demineralizer beds, and heaters, and then returns to the reactor.

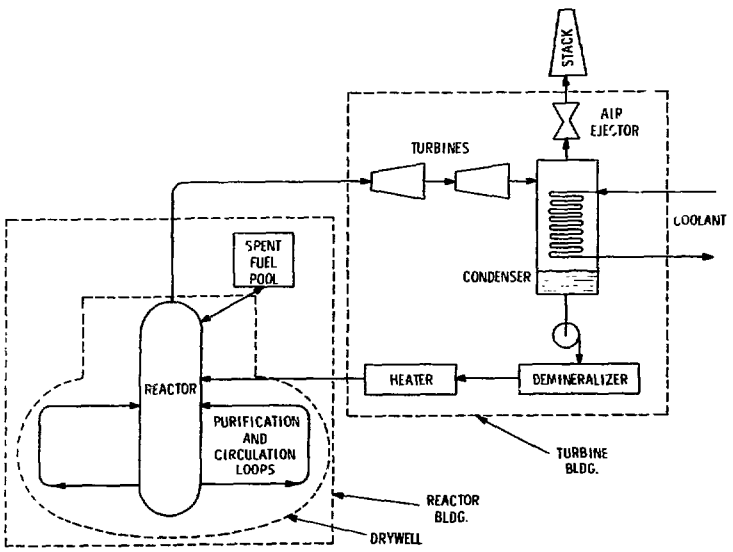

Fig. 1. BWR Coolant System

2. Gaseous Effluent Streams

Figure 2 is a simplified schematic diagram of the gaseous streams of a typical BWR. The most important gaseous effluent streams in which tritium and noble gases appear are the main condenser exhaust and the building ventilation exhausts (Smith).

*The core of a typical, modern BWR (Davis, Jacobson, WASH-1082) is an assembly of fuel elements consisting of Zircaloy-clad UO_2 and control rods of stainless steel-clad boron carbide (B_4C). The core and surrounding reactor vessel are located in an air-ventilated chamber (the drywell).

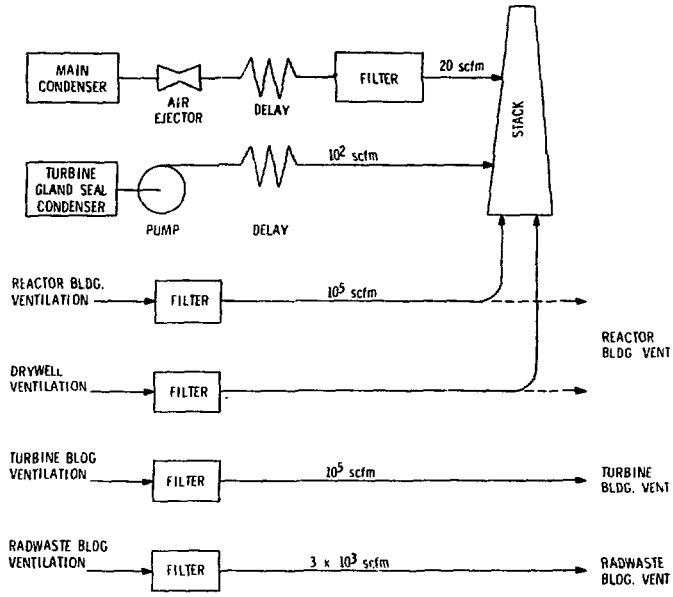

Fig. 2. Typical BWR Gaseous Radwaste System

The condenser is maintained at a partial vacuum, but air leaks into the system through seals. Some large BWR systems (see, for example, DOCKET-50342-3, DOCKET-50354-1) have been designed to accommodate an air-leak rate up to 60 cfm, but efforts are made to minimize the leak rate, and $\leq$ 20 cfm may be a more realistic value. This air and any other noncondensible gases are removed from the condenser system by a steam-jet ejector that vents to a condenser and the stack. In conventional plants this stream is delayed 20-30 min and then pumped through filters to the stack. The purpose of this holdup is mainly to permit sampling before discharge; the decay of some of the short-lived isotopes during holdup is an incidental benefit.

The turbine gland-seal exhaust is of secondary importance in the discharge of radioactive gases (Kahn, Smith). A large steam turbine will contain a number of gland seals to prevent steam leaking along the rotor (Etherington, Kearton). Seals of the labyrinth type usually consist of stationary circular blades, their planes positioned normal to the rotor axis. Process steam has customarily been supplied to these glands to prevent steam leakage from the ends of each turbine casing. Part of the steam supplied to the glands flows into the turbine, and the remainder flows into the gland exhaust system.

The gland exhaust stream is held up for 2-3 min to permit the decay of 7.13-sec ^{16}N and 26.8-sec ^{19}O, formed by neutron activation of air and water in the core. The main condenser exhaust and the gland-seal exhaust should carry the same radioactive constituents; however, only a small amount of the primary steam enters the gland-seal system and the release rate of radioactive gases from the gland-seal system to the environs will be approximately 0.1% of that from the main condenser off-gas system (Kahn).

Plans for operation of large BWRs of the near future (DOCKET-50354-1) include the use of a steam source that is independent of the primary cycle, thereby eliminating radioactive emissions from the gland-seal exhaust, even though this source is relatively small. The importance of the ventilation exhaust in the release of tritium at the reactor site is discussed in Section II.C.2.

3. Liquid Effluent Streams

Figure 3 is a simple schematic diagram of the liquid radwaste system of a typical 1000-MWe BWR, showing estimates of the flow rates of water. A major part of the total tritium that is released at the BWR site is expected to pass through this system. The typical BWR liquid radwaste scheme (Jason, Loy) involves segregation into two internal streams on the basis of radioactivity and dissolved-solids content (as indicated by ionic conductivity). "Clean" liquid waste (relatively high radioactivity and low conductivity), collected mainly from equipment drains, is filtered, demineralized, and recycled. "Dirty" liquid waste (relatively low radioactivity and high conductivity), collected mainly from floor drains, is unsuitable for demineralization because of its high dissolved-solids content, and is therefore filtered and discharged.

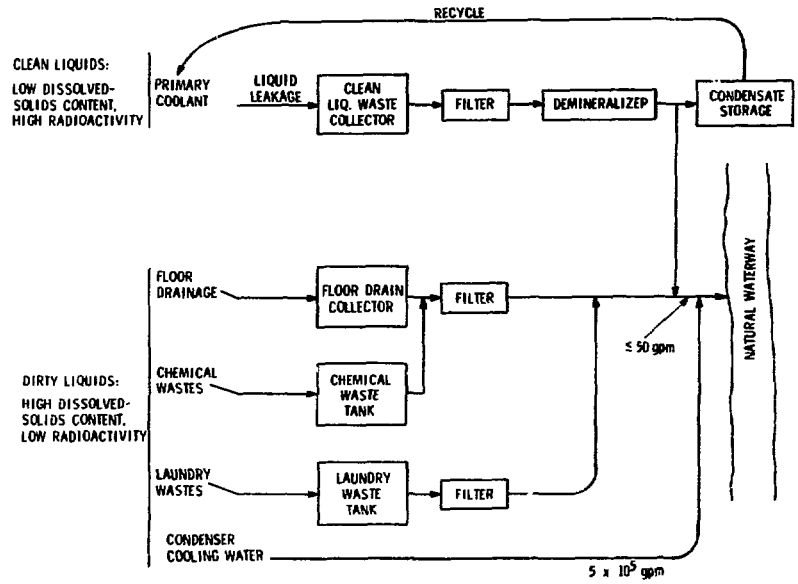

Fig. 3. Typical BWR Liquid Radwaste System

B. Generation of Tritium in Boiling Water Reactors

Tritium is generated in a BWR in several locations (Smith): (1) in the fuel (by light-particle formation in fission), (2) in control rods (by neutron activation of boron), (3) in the primary coolant (by neutron activation of

naturally-occurring deuterium), and (4) in structural materials (by neutron activation of lithium and boron impurities). The latter source is of minor importance. Expected rates of tritium generation in a 1000-MWe BWR are summarized in Table 1.

TABLE 1. Estimated Rate of Tritium Generation and Rate of Appearance in Waste Streams of a BWR

Source	Estimated Rate, Ci/1000 MWe-yr	
	Generation	Appearance in Waste Streams
Fission	2×10^4[a]	2×10^2[b]
Deuterium Activation, $^2H(n,\gamma)^3H$	14[c]	14
Boron Activation[d] (control rods)	10^4[c]	0

[a] Based on thermal fission yield of 1.3×10^{-4} tritons/fission, (Dudey) 2.7×10^{21} fissions/MWt-d (*i.e.*, 200 MeV/fission), and a thermal-to-electrical conversion efficiency of 33%.

[b] Assuming 1% defective cladding, about 200 Ci will appear in primary coolant and the same amount as an upper limit may appear in spent-fuel coolant (Section C.2.).

[c] Smith.

[d] $^{10}B(n,2\alpha)^3H$ and $^{10}B(n,\alpha)^7Li$; $^7Li(n,n\alpha)^3H$.

C. Appearance of Tritium in Boiling Water Reactor Waste Streams

1. Mechanisms and Amounts

All tritium that appears in the primary coolant of a BWR will eventually appear in effluent waste streams. The mechanisms of tritium appearance in the coolant include (1) formation in the coolant by neutron activation of deuterium, (2) escape from fuel rods through cladding defects, and (3) possible escape from control rods by permeation of the cladding or through cladding defects. Although tritium diffuses through stainless steel under certain conditions (Chaney), the extent of penetration of the stainless steel cladding of control rods at significant rates under the conditions of LWR operation has not been well defined (Kouts, Smith), and recent tests suggest that tritium would not escape from the B_4C matrix of control rods (Ebersole 1972). Therefore, although control rods contribute to the generation of tritium at the reactor site, they are not considered to contribute to the appearance of tritium in reactor waste streams.

It has been suggested (Ray) that tritium excaping through fuel-cladding defects as atomic or molecular tritium is converted rapidly to THO in the primary coolant because the conversion is promoted by the radiation field. This suggestion is consistent with experimental findings (Eakins, Yang) that the rate of conversion of tritium to tritiated water

is proportional to the activity in the system. Therefore, in current operations of the typical BWR, all tritium entering the primary coolant is expected to be discharged eventually to the environment as tritiated water in vapor or in liquid form.

Data obtained by monitoring for release of tritium in BWR liquid effluents have been used to calculate emissions in terms of Ci/1000 MWe-yr which are presented in Table 2. The values range from 10 to 1000 Ci/1000 MWe-yr, indicating that cladding failure in several instances have exceeded the expected values discussed below.

TABLE 2. Amounts of Tritium in Liquid Discharges at BWRs

Reactor	Cladding[a]	Ci/1000 MWe-yr			
		1969[b]	1970[b]	1971[c]	1972[c]
Dresden-1	Zr	61	31	122	325
Big Rock Point	Zr	590	1188	236	239
Humboldt Bay	Zr	114	141	190	302
LaCrosse	SS	-	1300	3900	4100
Nine Mile Point	Zr	-	46	36	73
Oyster Creek	Zr	128	55	48	120
Millstone 1	Zr	-	-	30	55
Dresden 2,3	Zr	-	-	123	90
Monticello	Zr	-	-	4.3	1.8×10^{-4}
Pilgrim 1	Zr	-	-	-	44[d]
Quad-Cities 1,2	Zr	-	-	-	10[d]
Vermont Yankee	Zr	-	-	-	0[d]

[a](NEI).

[b]Calculated from data in Logsden, 1971.

[c]Calculated from data in Nucleonics Week, Report on Releases at Radioactivity in Effluents Solid Waste from Nuclear Power Plants for 1972 (Directorate of Regulatory Operations, USAEC, August 1973).

[d]Plants operated less than 1 year.

Various design figures have been used in safety analyses (Denton) for the percentage of cladding failures expected, *e.g.*, 0.25, 0.5 and 1%; these values correspond to the release of about 50, 100 and 200 Ci/yr of fission-produced tritium, respectively, to the coolant of a 1000-MWe BWR.

On the other hand, one set of General Electric design figures indicates that a lesser amount, about 17 Ci/yr (Smith), of tritium is expected to appear in the reactor water of a 1100-MWe BWR; this value, however, implies that if 14 Ci/yr are formed by deuterium activation (Table 1), then only 3 Ci/yr of fission-product tritium enters the coolant through cladding defects (Table 1), a value that is small compared with observed releases in liquids (Table 2).

Another estimate, based on experience at the KRB Gundremmingen plant, a 237-MWe BWR, indicates that up to 1% of the tritium produced by fission might appear in either liquid or gaseous effluents (Smith). For a 1000-MWe BWR, this prediction implies that 200 Ci/yr could appear in gaseous or liquid streams.

Because the temperature of fuel assemblies is lower in the spent-fuel pool than in the reactor core, and also because a large fraction of any tritium available for leakage should have left the fuel during residence in the reactor, a leakage of much less than 200 Ci tritium/1000 MWe-yr into the spent-fuel pool is expected.

2. Pathways of Tritiated Water

Figure 4 is a schematic diagram of major paths for flow of tritiated water from internal sources to discharge in a BWR. The sources of tritiated water are the primary coolant and the spent-fuel coolant. Tritiated water moves from the primary coolant to effluent streams in equipment piping, through the main condenser exhaust, and through the gland-seal exhaust; it moves to effluent streams through routes external to equipment piping by leaks consisting of liquid flowing through the drains and the liquid waste-treatment system or vapor flowing through the containment ventilation streams. The latter route has been observed to be a major path in the Gundremmingen plant (Smith). Tritiated water moves from the spent-fuel pool to effluent streams through leaks to the reactor building and the liquid waste-treatment discharge.

Recent reports differ in identifying the main pathway for gaseous THO in a BWR. One report (Kent) identifies the exhausts from the main condenser and gland seal as the major carriers of gaseous THO (THO-H_2O vapors). A subsequent report (Smith) identifies the building ventilation streams that join in the stack as the major carriers of gaseous THO released at the reactor through numerous equipment leaks and by evaporation from pools of process water. If the latter hypothesis is true, the gas streams that must be considered in any tritium-retention process include the building ventilation streams. These streams will contain a large volume of air, a relatively large volume of atmospheric H_2O, and a low ratio of THO to H_2O. As an example, the stack exhaust of the KRB Gundremmingen plant carries about 100 tons of moisture per day, of which one tenth to one third is process water (Smith).

Surveillance at Dresden I reactor (Kahn) showed that, of all tritium discharged at the site, about 10% was in gaseous effluents and 90% was in liquid effluents. However, only the delay line of the main-condenser exhaust was monitored to obtain a measurement of the tritium content of gaseous effluents. The surveillance report recommended that the stack effluent be monitored in future work, from which it is inferred that the total THO

released in gases may have been greater than that in the condenser exhaust.

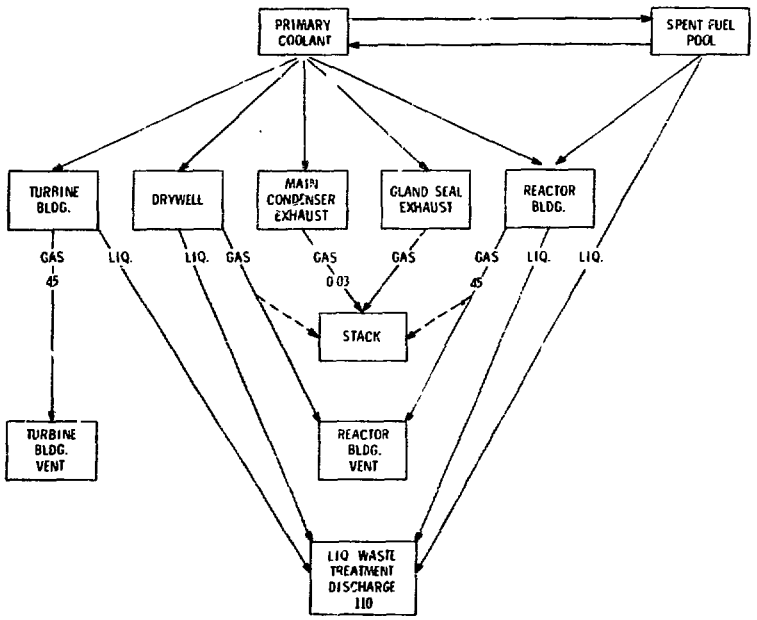

Fig. 4. Possible Paths of Tritiated Water in a BWR (estimated amounts in Ci ^{3}H/1000 MWe-yr, based on 1% fuel failure)

3. Effect of Advanced Management Schemes

It has been suggested that new modes of BWR management will tend to increase the recycle of primary coolant (Smith) to lessen tritium releases, but the extent of recycle that is possible seems as yet not to have been established. Thus, in future operation of large BWRs, the fraction of the total tritium released that will appear in the liquid streams will probably be smaller than for Dresden I (preceding section) because of efforts to increase recycling of primary coolant; perhaps 50% will be released in gaseous effluents and 50% in liquid effluents (Smith). Reactor monitoring data presently available are apparently not sufficiently detailed to give a very precise value for distribution of THO between liquid and gaseous effluents.

The retention of tritium within the plant during the entire life of the plant would require, in addition to coolant recycle, the elimination of leaks in all primary coolant conduits, the removal of all water vapor from the main condenser exhaust, and the removal of water vapor from the ventilation system of the spent-fuel storage pool.

4. Conclusions

The amount of tritium released at a 1000-MWe BWR site is expected to be about 200 Ci/1000 MWe-yr on the basis of an assumed cladding-failure level of 1%. The form of tritium released at a BWR is expected to be THO contained in effluent streams whose major constituents are liquid water or mixtures of water vapor and air. Estimates of the composition of major tritium-bearing effluents of a BWR are summarized in Table 3 based on estimates of the average concentration of tritium in the primary coolant and of

TABLE 3. Estimated Composition of Tritium-Bearing BWR Effluents (based on 1% fuel failure)

Type of Effluent	Constituent	Amounts/1000 MWe-yr		
		Ci	Liters(STP)	Metric Tons
Discharge from Liquid Waste Treatment[a]	THO	110	-	-
	H_2O(liquid)	-	-	10^5
Main Condenser Exhaust[b]	THO	0.028	-	-
	H_2O(vapor)	-	-	25
	Air	-	3×10^8	-
Reactor Building Ventilation[c,d]	THO	45	-	-
	H_2O(vapor)	-	-	10^5
	Air	-	2×10^{12}	-
Turbine Building Ventilation[c,d]	THO	45	-	-
	H_2O	-	-	10^5
	Air	-	2×10^{12}	-
TOTAL		200		

[a] Based on discharge of 50 gpm (DOCKET 50387-19) of liquid from waste treatment system (before dilution with condenser cooling water) and a tritium concentration of 1.1×10^{-3} μCi/ml in process water, consistent with the prediction (Smith) that the concentration range is 10^{-2} to 10^{-3} μCi/ml.

[b] Based on 20 scfm flow of air saturated with process water at 50°C and a concentration of tritium in liquid process water of 1.1×10^{-3} μCi/ml.

[c] Based on 10^5 scfm air flow saturated at 25°C with water of which 1/10 to 1/3 may be process water (Smith) with tritium concentration as assumed above.

[d] Somewhat smaller quantities of tritiated water released to ventilation streams can be estimated from predictions of leakages of primary steam into buildings and concentration of tritium in process water. Examples of predicted steam leaks to BWR reactor building are 360 lb/hr (Binford) and 480 lb/hr (Thomasson). If the tritium concentration of process water is that assumed above, then the amount of tritium released to a BWR reactor building ventilation would range up to about 4 Ci/1000 MWe-yr. Examples of predicted steam leaks to BWR turbine building range from 340 to 3000 lb/hr (Binford, DOCKET 50354-1, DOCKET 50387-19, Thomasson). For the tritium concentration in process water assumed above, the amount of tritium released to BWR turbine building ventilation would range up to 26 Ci/1000 MWe-yr.

flow rates of reactor streams. The data in Table 3 are consistent with the prediction (Smith) that, of the total tritium produced in fission, about 1% may appear at the BWR site with about half of it released in gaseous effluents, mainly in ventilation, and the other half in liquid effluents. The most difficult problems to be faced in any scheme for isolating tritium from BWR waste streams are related to the very large ratio of H_2O to THO and the large total volumes of waste.

D. Generation of ^{85}Kr in Boiling Water Reactors

The noble gases xenon and krypton are generated in significant quantities by fission within the fuel assemblies and in relatively insignificant quantities by fission of tramp uranium on the surface of fuel assemblies. Because of their short half-lives, the generation rates of most of the noble-gas fission-product isotopes are inconsequential in relation to long-term disposal problems. The generation rate of ^{85}Kr for 1000 MWe-yr of power generation by thermal fission in light-water reactors is 4×10^5 Ci/1000 MWe-yr, assuming that the ^{85}Kr yield from thermal fission of ^{235}U is 0.27% (Katcoff), that 200 MeV of energy is released per fission [*i.e.*, 2.7×10^{21} fissions occur per Mwdt], and that the thermal efficiency of the BWR is 33%.

E. Appearance of ^{85}Kr in Boiling Water Reactor Waste Streams

1. Mechanisms and Amounts

Except for the relatively insignificant amounts of noble gases released by the fission of tramp uranium, noble gases are emitted at the reactor site by diffusion out of the oxide matrix of the fuel, through cladding defects into the coolant, and thence into the main condenser where the steam-jet ejector sweeps out noncondensible gases. The amounts of noble gases appearing in waste streams depend on the extent of fuel failure and also, for short-lived isotopes, on the time required for emergence from the oxide matrix and cladding, *i.e.*, on the leak rate. Three models have been defined (Blomeke, Kent) to provide a means for estimating the composition of noble-gas mixtures emerging from the fuel under various conditions: (1) a recoil mixture, representing the noble gases released from the fuel by recoil soon after fission, (2) an equilibrium mixture, representing the noble gases present in the fuel after one month of irradiation (at this time, all isotopes except ^{85}Kr are in radioactive equilibrium); and (3) a diffusion mixture, representing the gases that diffuse out of the fuel matrix, the composition of which depends on the assumed rate of diffusion and the half-lives of the isotopes. The composition of gases emerging from defective cladding will therefore correspond to one or a combination of these models, depending on the irradiation time and the rate of escape from fuel. Calculations of the composition of noble-gas fission-product mixtures as a function of post-irradiation decay time (Blomeke) indicate that ^{85}Kr is a minor constituent at short decay times, but becomes a major constituent after decay times of 3 to 60 days.

Estimates of the amounts of noble gases emitted at the site of a 1000-MWe BWR have been quoted frequently for plant safety-analysis reports

on the basis of emission of a diffusion mixture after 30-min holdup at an arbitrary rate of 10^5 μCi/sec, *i.e.*, 3 x 10^6 Ci/yr (Table 4). The reports state that emissions during operation are expected to be at 2% of these rates or 6 x 10^4 Ci/yr, and that the highest observed emissions to 1969 are about 20% of the estimated rates (DOCKET 50342-5, DOCKET 50354-1). Emissions in 1970 of total noble gases at Oyster Creek (USA), 542 MWe; Tarapur (India) 1 & 2, 420 MWe; and Tsuruga (Japan), 331 MWe, ranged from 2 x 10^4 to 8 x 10^5 Ci/yr, (Kent), a range that falls below the total rate of 3 x 10^6 Ci/yr for the hypothetical 1000-MWe BWR listed in Table 4, but exceeds the expected 2 to 20% of design value (Table 4 footnotes). Published sums of noble and activation gases (Table 5) released at reactor sites (Logsdon, Logsdon 1971, WASH-1198) are not easily translated into estimates of release of noble gases alone (Table 5). Most of the ^{85}Kr leaving the BWR follows the path from the primary coolant to the main condenser exhaust and from there to the stack or building vent. Lesser amounts from the primary coolant reach the stack or vent by way of leaks into ventilated spaces or the gland-seal exhaust, if sealed by process steam. Only 0.1% of the noble gases follow the latter route (Kahn). Any ^{85}Kr released from the spent-fuel pool reaches the stack or vent through the pool ventilation system.

TABLE 4. Design Estimates[a] of Radioactive Noble-Gas Isotopes Emitted Annually by 1100-MWe BWR

Isotope	Half-Life	Ci/yr	mol/yr[b]
^{89}Kr	3.2 m	1.1 x 10^4	1.9 x 10^{-7}
^{137}Xe	3.8 m	2.9 x 10^4	6.0 x 10^{-7}
^{135m}Xe	15.6 m	2.7 x 10^5	2.2 x 10^{-5}
^{138}Xe	14 m	8.2 x 10^5	7.4 x 10^{-5}
^{87}Kr	76 m	4.92 x 10^5	2.0 x 10^{-4}
^{83m}Kr	1.86 h	9.5 x 10^4	5.5 x 10^{-5}
^{88}Kr	2.8 h	5.4 x 10^5	4.8 x 10^{-4}
^{85m}Kr	4.4 h	1.6 x 10^5	2.3 x 10^{-4}
^{135}Xe	9.2 h	5.77 x 10^5	1.7 x 10^{-3}
^{133m}Xe	2.3 d	6.3 x 10^3	1.1 x 10^{-4}
^{133}Xe	5.27 d	1.7 x 10^5	6.9 x 10^{-3}
^{131m}Xe	11.9 d	3.8 x 10^2	3.5 x 10^{-5}
^{85}Kr	10.76 y	2.6 x 10^2	7.8 x 10^{-3}

[a]Calculated from design values for a diffusion mixture, based on a total emission rate of 10^5 μCi/sec (3 x 10^6 Ci/yr) after 30-min holdup. Operating values may be 2% of these observed maximum values (to 1969) and in smaller BWRs, they were 20% of these (DOCKET 50342-5, DOCKET 50354-1). These design estimates are apparently based on a fuel failure level of less than 1%; compare 2.6 x 10^2 Ci/yr with 4 x 10^2 Ci/yr for ^{85}Kr emission at 1% failure.

[b]Calculated from Ci/yr and specific activities, amounts in terms of mol/yr are included to facilitate estimation of weights or volumes.

TABLE 5. Observed Release of Noble and Activation Gases at BWRs

Plant	Ci/1000 MWe-yr			
	1969[a]	1970[a]	1971[b]	1972[b]
Oyster Creek	1.8×10^5	2.7×10^5	1.2×10^6	1.7×10^6
Nine Mile Point	6.8×10^3	4.4×10^4	7.3×10^5	1.4×10^6
Millstone-1	---	---	6.5×10^5	1.9×10^6
Dresden-1	8.7×10^6	6.0×10^6	1.0×10^7	6.6×10^6
Dresden-2 & 3	---	---	1.8×10^6	4.8×10^5
LaCrosse	5.5×10^4	4.7×10^4	2.3×10^4	1.0×10^6
Monticello	---	---	5.4×10^5	1.8×10^6
Big Rock Point	4.2×10^6	6.1×10^6	6.5×10^6	5.9×10^6
Humbolt Bay	1.1×10^7	1.1×10^7	1.3×10^7	1.0×10^7
Pilgrim	---	---	---	1.9×10^5[c]
Quad Cities-1 & 2	---	---	---	2.8×10^5
Vermont Yankee	---	---	---	1.1×10^6[c]
TOTALS	2.4×10^7	2.3×10^7	3.4×10^7	3.2×10^7
AVERAGE	4×10^6	4×10^6	4×10^6	3×10^6

[a] Normalized to 1000 MWe-yr from data in Logsdon 1971 and includes activation gases.

[b] Normalized to 1000 MWe-yr from data in Nucleonics Week and Radioactivity in Effluents & Solid Waste from Nuclear Power Plants for 1972, Directorate of Regulatory Operations, USAEC, August 1973 (DRO).

[c] Plants operated less than one year.

2. Pathways for ^{85}Kr

Recently, several computer models have been developed (Binford, Jauho, PGE) that attempt to describe reactor releases of gaseous fission products for a range of operating conditions. Figure 5 is a much simpler representation, *i.e.*, a schematic diagram of the main pathways for ^{85}Kr in a BWR.

3. Composition of Main Condenser Exhaust

If separation of noble gases from effluents of a BWR were to be required, the exhaust from the main condenser is the stream that will receive primary attention, because it contains about 99.9% of all noble gases released into the primary coolant and contains smaller amounts of air than the stack exhaust or building vent.

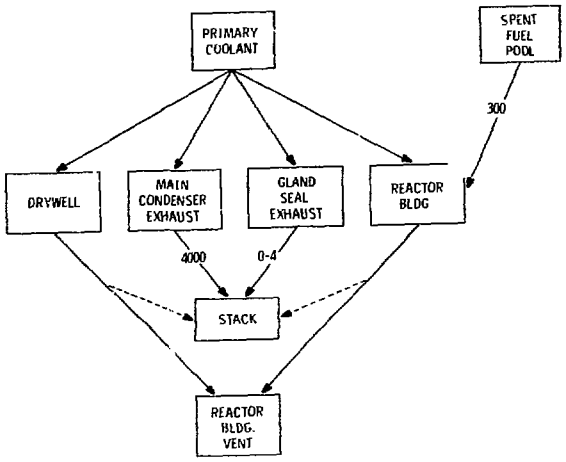

Fig. 5. Major Pathways of ^{85}Kr in a BWR (estimated amounts in Ci ^{85}Kr/1000 MWe-yr based on 1% cladding failure)

Table 6 shows the predicted total annual amounts of xenon and krypton in the main condenser exhaust stream of an 1100-MWe BWR having an air in-leakage of 20 cfm. Since the amounts of fission-product gases are small compared with the amount of air leakage, the concentration of xenon and krypton in the exhaust stream are not significantly higher than the normal concentrations of xenon and krypton in air, namely, 0.087 and 1.14 ppm respectively (Weast).

4. Effects of Advanced Management Schemes

Proposals of advanced management schemes that would affect noble-gas-bearing effluents in BWRs include the use of catalytic recombiners and treatment of main condenser exhaust by cryogenic traps or by ambient-temperature charcoal beds (Denton). The catalytic recombiner removes H_2 and O_2 from the noble-gas bearing stream. Some schemes, including treatment of main condenser exhaust by absorption on charcoal traps at ambient or cryogenic temperature or by condensation at cryogenic temperature, are intended only for temporary hold-up to permit decay of short-lived xenon and krypton isotopes and activation gases. A cryogenic condensation after delay can also be added if ^{85}Kr is to be collected for ultimate storage (Michels, Winters).

5. Conclusions

If cladding failure occurs at the level of 1%, then 4×10^3 Ci/1000 MWe-yr will enter primary coolant and be swept out through the main condenser exhaust. Other constituents of this stream include water vapor, H_2 and O_2 from radiolysis of primary coolant, and air. The ratio of xenon to krypton in the exhaust stream will be about the same as in air, with an estimated total of about 300 liters (STP)/1000 MWe-yr of stable krypton and about 30 liters (STP)/1000 MWe-yr of stable xenon. Several schemes proposed for advanced

management would alter the composition of the main condenser exhaust: (1) the H_2 and O_2 would be eliminated through the use of a catalytic recombiner and (2) the short-lived isotopes of Kr, Xe, N, and O would be eliminated by decay during temporary holdup on charcoal at ambient or low temperature, or by condensation at cryogenic temperatures. For 1% cladding failure, an additional $\geq$300 Ci/1000 MWe-yr of ^{85}Kr might appear in the coolant of the spent-fuel pool and be swept into the ventilation of the reactor building.

TABLE 6. Major Constituents of Main ^{85}Kr-Bearing Effluent (Main Condenser Exhaust) of BWR[a]

Constituent	Amounts/1000 MWe-yr		
	Ci	Liters(STP)	Metric Tons
^{85}Kr	4×10^{3}[b]	3×10^{-1}	
Air[c]		3×10^{8}	
H_2O Vapor[c]			25
Total Stable Kr[c,d]		3×10^{2}	
Total Stable Xe[c,d]		3×10^{1}	
H_2[e]		2×10^{9}	
O_2[e]		1×10^{9}	

[a]Short-lived isotopes of Xe and Kr and activation gases not listed.

[b]Assuming 1% fuel cladding failure.

[c]On basis of 20 scfm air leak and condenser temperature of about 50°C (DOCKET 50387-19).

[d]Fission products plus contribution from air leakage.

[e]DOCKET 50352-5, Limerick. These gases would be eliminated in advanced reactor management systems by a catalytic recombiner.

III. BEHAVIOR OF TRITIUM AND ^{85}Kr IN PRESSURIZED WATER REACTORS

A. Pressurized Water Reactor System Descriptions

1. Coolant Cycle

The primary coolant in a pressurized water reactor (PWR) is not the working fluid, as it is in the direct-cycle BWR. Rather, in a PWR, heat is transferred from the primary coolant to the working fluid (secondary coolant) in a steam generator (Davis, Loose, WASH-1082) that is located in the same containment as the core* (Fig. 6).

*The typical PWR core is assumed to consist of sintered UO_2 pellets clad in Zircaloy (WASH-1082); the stainless steel clad fuels of early PWRs are being replaced by Zircaloy-clad fuels.

The primary coolant system includes a pressurizer that maintains the water system in a nonboiling condition at the operating temperature, recirculating pumps, and the reactor-coolant side of the heat exchanger. The primary coolant, contained in a sealed loop at a pressure of 2250 psia, may have a cover gas of nitrogen to exclude oxygen from the system or a partial pressure of hydrogen may be maintained in the system to force recombination with the oxygen formed by radiolysis of primary coolant. A side stream of the primary coolant is continually bled off into a subsystem (primary coolant letdown*), usually located in an auxiliary building, for control of chemical content and volume, for coolant purification, and for the treatment and disposal of both gaseous and liquid wastes.

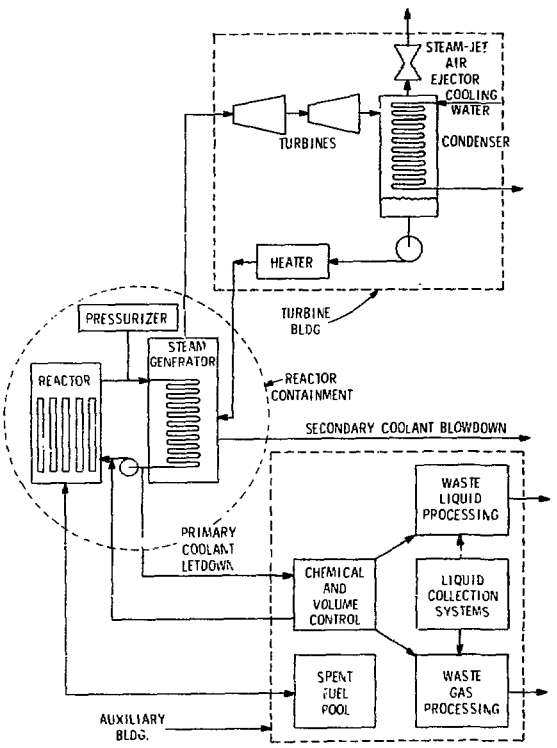

Fig. 6. Typical PWR System

*Flow of coolant from reactor for pressure adjustment, boron concentration control, sample collection, or purification.

Reactivity control in PWRs has customarily been effected, in part, by regulating the concentration of boron dissolved in primary coolant. In a conventional chemical shim control system, the boron content of the coolant water is adjusted through first removing water by evaporation and then either diluting with demineralized water or concentrating with boric acid. In an advanced PWR concept, the concentration of boron in the coolant is regulated by adjusting the temperature of an ion-exchange resin to the levels where boron is either absorbed from or released to the coolant (Loose).

The secondary coolant absorbs heat at the heat exchanger, forming steam which flows through the turbine, condenses in the condenser, and returns through pumps to the heat exchanger. The turbine and condenser are customarily located in a separate turbine building. The condenser is maintained at a partial vacuum by a steam-jet ejector that removes the air that leaks into the secondary coolant system. The secondary coolant system also contains a subsystem for adding fresh water and discharging (blowdown) recycled water to reduce the accumulation of dissolved solids. It is current practice to cool the main condenser with a flow of water diverted from a nearby natural waterway at a rate of about 8×10^5 gpm for a 1000-MWe PWR (Dierman).

2. Gaseous Effluent Streams

The typical PWR gaseous radwaste systems (Blomeke, Jason, Kahn 1971, Leonard, Wright) consist of streams that carry (1) the gases periodically discharged from the primary coolant system (this stream includes a compressor, tanks for holdup and decay of short-lived isotopes, and filters to remove particulate matter), (2) the main condenser exhaust, (3) the vapor from blowdown of the secondary coolant, and (4) reactor-building ventilation flow, including periodic (probably four times per year) containment purge and the ventilation flow from the turbine building and auxiliary building. The PWR gaseous radwaste streams are diagrammed in Fig. 7.

3. Liquid Effluent Streams

The simplest PWR liquid radwaste schemes have consisted of collecting all liquid wastes and processing them through a single evaporator. The evaporator bottoms are converted to solids with cement and drummed. The evaporator overheads are usually discharged into the condenser cooling-water effluent. Other schemes consist of segregation (Blomeke, Leonard) into "clean" wastes (mainly primary coolant letdown) and "dirty" wastes (collected from various building drains), processing of the two streams through separate evaporators, and recovery of the boric acid from the "clean" wastes (Fig. 8). PWR liquid wastes have also been segregated on the basis of hydrogen-containing deaerated wastes, originating from primary coolant, and aerated wastes, originating from secondary coolant and miscellaneous drains (Denton).

B. Generation of Tritium in Pressurized Water Reactors

Tritium is generated in PWRs by a number of mechanisms (Oestmann). The most important for the problem of tritium control are fission and neutron activation of boron solute in the primary coolant (Kahn 1971)

1. Fission

If the yield of tritium in thermal fission is assumed to be 1.3×10^{-4} tritons per fission (Dudey), light-water reactors (BWR and PWR) should produce about 18,500 Ci/1000 MWe-yr, equivalent to about 1.8 g or 0.6 mol/yr.

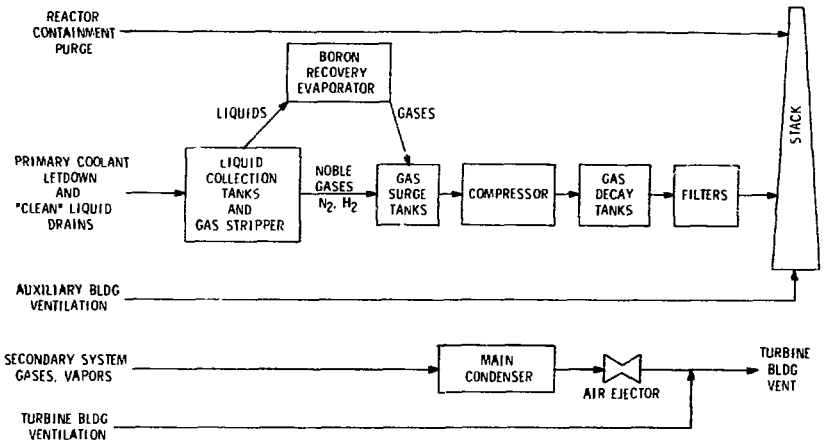

Fig. 7. Typical Gaseous Radwaste System for a Conventional PWR

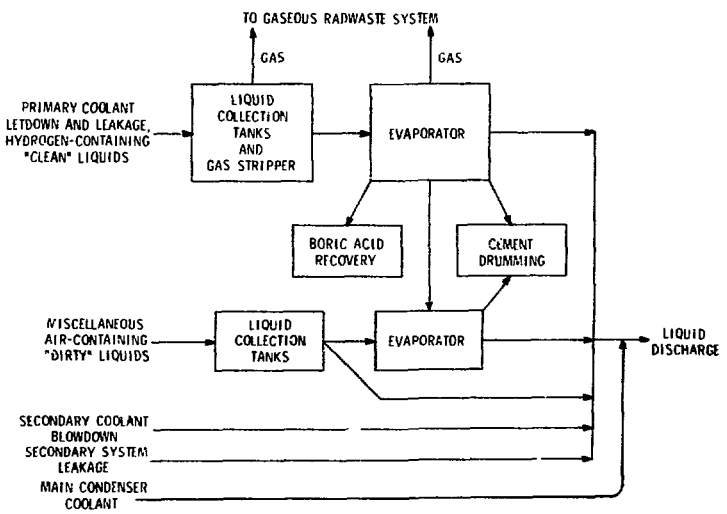

Fig. 8. Typical Liquid Radwaste System for a Conventional PWR (based on segregation of H_2-containing and air-containing streams to gaseous radwaste system)

The yield of tritium in fission is a function of both the neutron energy and identity of the fissioning nuclei; the thermal neutron yield from ^{239}Pu exceeds that from ^{235}U by a factor of about 2. As burnup increases, the fraction of fissions from ^{239}Pu and hence the rate of generation of tritium increases slightly.

2. Boron Shim

An additional 500 to 1000 Ci/1000 MWe-yr may be produced by neutron activation of boron in the primary coolant of a PWR (Kouts). The activation of ^{10}B by two mechanisms are considered:

(1) The fast-neutron reaction $^{10}B(n,2\alpha)^{3}H$.

(2) The two-step reaction $^{10}B(n,\alpha)^{7}Li(n,n\alpha)^{3}H$, which is estimated to produce only a small amount of tritium compared with that produced in the first reaction. The amount may become of importance if ^{7}Li is allowed to accumulate in the coolant (Kahn 1971, Ray).

3. Other Sources

Other neutron-activation sources of tritium in a PWR (Locante) involve ^{10}B in stainless-steel clad control rods, ^{6}Li and ^{7}Li in the coolant, and the naturally-occurring deuterium in the coolant.

Tritium production in a PWR from fixed, burnable-poison rods (containing B_2O_3) will be important only during life of a first core when reactivity holddown is required (Kouts). Subsequent loadings will not have poison-rod components. Control rods of silver-indium-cadmium alloy are to be used in modern PWRs; these metals do not produce tritium under neutron irradiation. Nearly all the large PWRs under construction specify Ag-In-Cd for control (NEI).

In Westinghouse PWRs, lithium is employed for pH adjustment of the reactor coolant. A maximum level of 2.2 ppm lithium is maintained by adding $^{7}LiOH$ or by using a cation-exchange demineralizer to remove any excess lithium that might arise from the $^{10}B(n,\alpha)^{7}Li$ reaction (Locante 1973). The use of 99.9% ^{7}Li in both the LiOH additive and the ion-exchange resins limits the amounts of ^{6}Li in the coolant and thus the $^{6}Li(n,\alpha)^{3}H$ reaction which has a relatively high thermal cross section (see Table 7).

C. Appearance of Tritium in Pressurized Water Reactor Waste Streams

1. Mechanisms and Amounts

Of the various mechanisms listed in Table 7, those most important to the appearance of tritium at the PWR site are (1) release of fission-product tritium through fuel-cladding defects and (2) neutron activation of boron dissolved in the coolant.

Since the mechanisms of appearance of tritium in PWR waste streams involve contact with primary coolant, it has been assumed that tritium would be present in these streams mainly as THO. Monitoring of the effluents of the Yankee-Rowe reactor (Kahn 1971) has shown that a very high fraction of tritium released at the site does indeed appear mainly in aqueous liquid waste (Fig. 9). The small fractions of total tritium that appear in gaseous waste streams, however, show a high ratio of elemental tritium to tritiated water vapor.

TABLE 7. Estimated Rate of Tritium Generation and Rate of Appearance in Waste Streams of a PWR

Source	Estimated Rate, Ci tritium/1000 MWe-yr	
	Generation	Appearance in Waste Streams
Fission	2×10^{4}[a]	2×10^{2}[b]
Burnable Poison Rods	1.2×10^{3}[c,d]	1.2×10^{3}[d]
Control Rods	0[c,e]	0
Soluble Poison Boron	$5\text{-}10 \times 10^{2}$[f]	$5\text{-}10 \times 10^{2}$[f]
^{7}Li Activation	14[c]	14
^{6}Li Activation	8[c]	8
Deuterium Activation	1[c]	1

[a]Based on thermal fission yield at 1.3×10^{-4} tritons/fission (Dudey), 2.7×10^{21} fissions/MWtd (*i.e.*, 200 MeV/fission), and a thermal-to-electrical conversion efficiency of 33%.

[b]Assumes 1% cladding failure.

[c]Data (Locante) converted to Ci/1000 MWe-yr.

[d]In initial cycle only.

[e]Control by Ag-In-Cd alloy.

[f]From data of Kouts.

Of the tritium generated by various sources (Table 7), that generated from sources dissolved in or in contact with primary coolant, *i.e.*, that formed by neutron activation of dissolved boron shim, lithium, and deuterium, is certain to appear in reactor effluents.

The amount of fission-product tritium appearing at the reactor, however, varies with the cladding integrity. Design values for fission-product appearance have been based on various percentages of Zircaloy cladding failure, *e.g.*, 0.1% (DOCKET 50382-30), 0.25% (Thomasson) and 0.2% (Wright). More recent experience has shown that in a given group of PWRs, the maximum Zircaloy cladding failure was 0.7% (Wright). Although as much as 80% of the fission-product tritium had been observed to permeate stainless steel cladding, early experience with diffusion of tritium through Zircaloy cladding in PWRs suggests

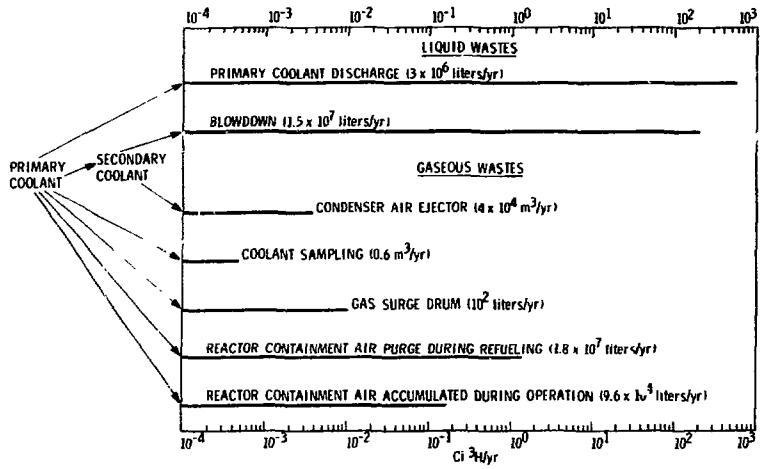

Fig. 9. Distribution of Tritium in Yankee-Rowe PWR System (Kahn 1971)

that about 1% of fission-product tritium might be expected to appear in effluent streams (Locante).

For this discussion, cladding failure will be assumed to occur at a rate of 1%; thus, the amount of fission-product tritium expected to appear in waste streams would be about 200 Ci/1000 MWe-yr. In addition, as much as 1000 Ci ^{3}H/1000 MWe-yr may appear as a result of boron activation; hence, the total amount of tritium appearing at the PWR site will be assumed to be 1200 Ci/1000 MWe-yr.

Data collected by the Environmental Protection Agency (Logsdon, Logsdon 1971) and the USAEC Directorate of Regulatory Operations (DRO) on amounts of tritium discharged in liquids from PWRs have been normalized to Ci/1000 MWe-yr, and the results are shown in Table 8. A comparison of these data shows that tritium discharge from PWRs with stainless steel-clad fuel is considerably higher than the estimated 1200 Ci/1000 MWe-yr and is one to two orders of magnitude higher than the tritium discharge from reactors using Zircaloy-clad fuel (with the exception of Yankee-Rowe). This is an indication of either greater diffusion of tritium through stainless steel or a higher percentage of failure for stainless steel cladding. Future monitoring at PWR sites will probably show smaller tritium discharges as stainless steel cladding is replaced by Zircaloy.

2. *Pathways of Tritiated Water*

Since all tritium released at a PWR site passes through primary coolant, the pathways of tritium are assumed to be those available to primary coolant. The tritium distribution has been calculated from an assumed concentration of tritium in the primary coolant and assumed leak rates of primary coolant through primary system defects into the reactor containment and the auxiliary building as well as through steam generator defects into the secondary coolant system.

TABLE 8. Tritium Found in PWR Liquid Discharges

Plant	Fuel Cladding[a]	Tritium Released in Liquids, Ci/1000 MWe-yr			
		1969[b]	1970[b]	1971[c]	1972[c]
Shippingport	Zr	517	38	NA	NA
Yankee-Rowe	SS-Zr[d]	8,900	3,100	10,000	10,200
Indian Point-1	SS	5,300	-	4,800	4,050
San Onofre-1	SS	11,000	13,000	11,800	10,400
Connecticut Yankee	SS	11,800	16,600	11,800	11,400
Robert Ginna	Zr	74	398	477	400
Point Beach 1 & 2	Zr	-	-	690	1,500
Maine Yankee	Zr	-	-	-	174[e]
Palisades	Zr	-	-	-	960
Robinson	Zr	-	-	420	685
Surry-1	Zr	-	-	-	100[e]

[a](NEI).

[b]Calculated from data tabulated by the Environmental Protection Agency (Logsdon 1971).

[c]Calculated from power production (Nucleonics Week) and release data (DRD).

[d]SS through 1970, Zr beyond.

[e]Plants operated less than one year.

The published estimates of leak rates vary widely (Table 9). For the present calculations of tritium transport resulting from primary coolant leaks, and average of the leak rates quoted by Binford and by Thomasson has been assumed. The concentration of tritium in primary coolant will vary with the size of leaks (Burns) and with coolant processing; however, an average value of 3×4^{-4} Ci/liter has been assumed for the present calculations. This concentration was derived from the assumption that 1200 Ci ^{3}H/yr is introduced into 3.5×10^{6} liter of water, the amount of primary coolant that may be processed and discharged per year from a 1000-MWe PWR (DOCKET 50382-30). The distribution of PWR effluents and tritium values are shown in Table 10, corresponding to the assumed operational conditions outlined in the footnotes of the table. These assumptions were also used to diagram tritium-transport in PWR (Fig. 10).

3. Effect of Advanced Management Schemes

Proposals for management of radwaste in PWRs to reduce the amounts of radioactivity released include segregation of tritiated and non-tritiated streams and recycle of the tritiated streams, as shown in Fig. 11. It has been

TABLE 9. Leakage Rates for Estimated Distribution of Tritium from PWRs

Leakage of Primary Coolant, lb/day			Secondary System Leakage, lb/day	Blowdown Rate lb/day	Reactor Rating, MWe	Ref.
To Containment	To Secondary System	To Auxiliary Building				
50	-	-	-	-	∿1100	Locante 1973
30	167	-	167	300,000	∿600	Wright
<1200	1020	84	negligible	10,000	∿1100	Docket 50382-30
240	120	174	-	120,000	-	Binford
330	167	167	-	120,000	∿1100	Thomasson

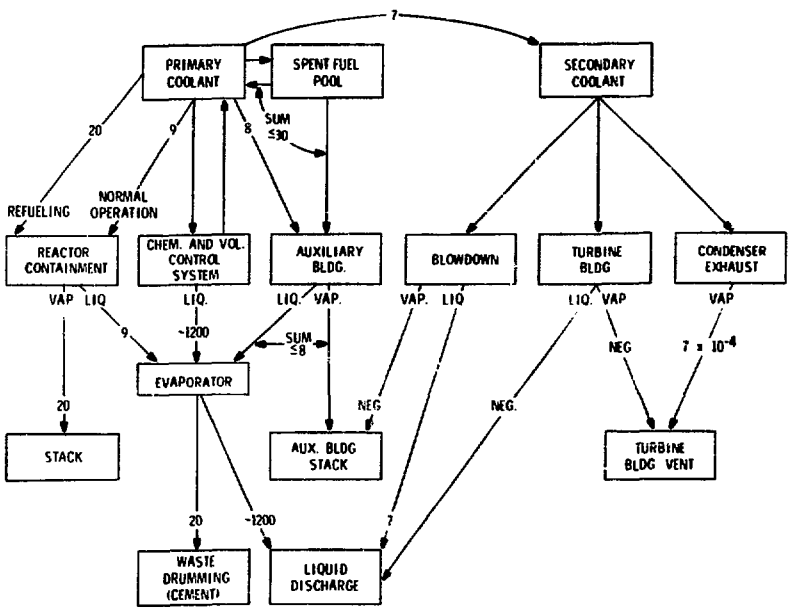

Fig. 10. Transport of Tritium in a Conventional 1000/MWe PWR (amounts in Ci H/1000-MWe-yr based on assumptions outlined in footnotes of Table 10; NEG. = negligible amount)

suggested that tritium released to primary coolant might thus be retained in the primary coolant system for the life of the plant (Gallagher, Loose). Although segregation and recycle of tritiated water seems feasible, total containment of tritium would imply that all leakages would have to be eliminated or collected and retained. If no tritium leaked from the primary system, at

TABLE 10. Estimated Composition of Tritium-Bearing Effluents of PWRs

Effluent	Constituent	Amounts/1000 MWe-yr		
		Ci	Liters(STP)	Metric Tons
Primary Coolant Letdown (discharge from chemical volume control system)	THO	1200[a]	-	-
	H_2O	-	-	3×10^{3}[b]
Evaporator Bottoms (drummed cement)	THO	20[c]	-	-
	H_2O	-	-	60[c]
Secondary Coolant Blowdown	THO	7[d]	-	-
	H_2O	-	-	2×10^{4}[e]
Auxiliary Building Ventilation	THO	8[f]	-	-
Containment	THO	20[g]	-	-
Atmosphere	H_2O	-	-	2.3×10^{3}[h]
Purge	Air	-	10^{11}[h]	-
Main Condenser Exhaust	THO	7×10^{-4}[i]	-	-
	H_2O	-	-	2[j]
	Air	-	3×10^{8}[j]	-

[a]Assumes 1% of fission product tritium (*i.e.*, 200 Ci ^{3}H/1000 MWe-yr leaks through cladding and that 10^{3} Ci ^{3}H/1000 MWe-yr is formed by neutron activation of boron in primary coolant.

[b]Based on estimated discharge of 780,000 gal/yr of liquid wastes from the boron management system of Waterford Unit No. 3 (DOCKET 50382-30).

[c]Based on 15,600 gal/yr of evaporator bottoms from boron management system (DOCKET 50382-30) and 3×10^{-4} Ci/liter concentration of tritium.

[d]Based on leakage of 144 lb/day of primary coolant containing 3×10^{-4} Ci/liter of tritium into secondary system and exit of essentially all tritium in secondary system through blowdown (see footnote j).

[e]Based on blowdown rate of 120,000 lb/day.

[f]Assuming leakage of 170 lb/day of primary coolant containing 3×10^{-4} Ci/liter of tritium into auxiliary building. The partition of 8 Ci of tritium between liquid that flows to waste treatment system and ventilation streams is undefined.

[g]Assuming that the ratio of tritiated water released in containment purge (20 Ci) to total tritium released at the reactor is about the same as that for Yankee-Rowe (*e.g.*, 13/800). Also assuming most of the 9 Ci of tritium leaking to containment in normal operation (285 lb/day of water containing 3×10^{-4} Ci/liter) goes to liquid drains (see Fig. 10).

[h]Assuming the containment purge of 80,000 scfm of air for 30 days/yr is saturated with H_2O at 25°C.

[i]Assuming that 7 Ci of tritium transferred to the secondary system as THO is divided proportionately between 2 metric ton of water vapor/1000 MWe-yr in condenser exhaust and 2×10^{4} metric ton of liquid water/1000 MWe-yr in blowdown.

[j]Based on water saturation at 50°C of 20 scfm air flow.

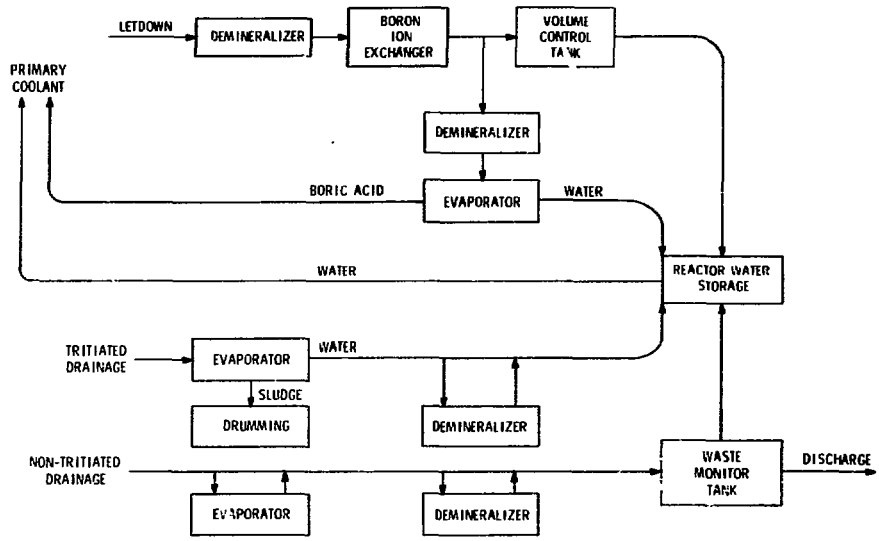

Fig. 11. Advanced PWR Liquid Radwaste Scheme (based on segregation of tritiated and non-tritiated streams; conventional scheme shown in Fig. 8)

the end of 40 years of operation, the tritium inventory (taking into account the decay of about one-third of the amount formed) would be about 3×10^4 Ci contained in the primary-coolant pool and refueling-water storage tank ($\sim 2 \times 10^3$ metric tons). If leaking of the primary system occurs, and the leakages are collected and retained, the volume accumulated over the life of the plant might be much greater.

Advanced management has also included proposals to treat secondary coolant blowdown by demineralization and filtration (Wright). This treatment would effectively remove fission and activation products in ionic and particulate forms but would not remove tritium appearing in the secondary system as a result of steam generator defects.

4. Conclusions

The most important mechanisms of tritium appearance in PWR effluents are neutron activation of boron dissolved in the primary coolant and the escape of fission-product tritium through cladding defects into the primary coolant. Tritium released to the coolant appears mainly in the form of THO and follows liquid pathways available to primary coolant. Leaks in the primary coolant system result in discharges of minor fractions of the total tritium to ventilation and liquid drains, to containment, as vapor to main condenser exhaust, and as liquid to secondary coolant blowdown. The major portion of tritium discharged from the typical 1000-MWe PWR appears in liquid discharges during primary coolant letdown, possibly 1.2×10^2 Ci in 3×10^6 liters annually.

Advanced PWR management proposals would segregate tritiated and non-tritiated plant fluids aiming at total recycle of tritiated fluids. If this proves successful, the inventory of a 1000-MWe plant at decommissioning, after about 40 yr of operation, would be a total of about 3×10^4 Ci of tritium contained in 2×10^3 metric tons of primary coolant and refueling water (concentration, 15 μCi/ml). This burden, normalized to electric power, would be only (500 Ci tritium in 50 metric tons H_2O)/1000 MWe-yr.

A minor fraction of fission-produced tritium may also be released into the spent-fuel pool. This tritium, as THO, is expected to appear partly as vapor in the ventilation stream of the fuel pool but probably mainly as liquid in effluent from the spent fuel pool. For the reasons listed in Section II. C.1, the amounts of tritium released by failed spent fuel are expected to be small relative to the amounts released by failed fuel in the core. However, because of the contact with reactor water in the transfer of fuel from core to storage, the concentration of tritium in the spent-fuel pool is expected to approach that of the primary coolant.

D. Generation of ^{85}Kr and Appearance in Pressurized Water Reactor Waste Streams

The only source of ^{85}Kr in a PWR is fission; hence, the generation rate for a PWR should be about the same as that estimated for a BWR (Section II.D.) *i.e.*, 4×10^5 Ci/1000 MWe-yr.

1. Mechanisms and Amounts

Since the primary coolant in the PWR system is opened infrequently, much of the short-lived activity decays before venting, and the appearance of short-lived noble-gas isotopes and activation gases at the site will be much less than in the case of the direct-cycle BWR, in which gases introduced into the primary coolant are promptly swept out through the main condenser exhaust. In the conventional PWR, short-lived noble-gas isotopes and activation gases will appear mainly in water leaked from the primary coolant system, rather than in gases obtained from secondary-coolant-stripping operations.

The mechanism of appearance of ^{85}Kr in PWR effluent streams starts with its diffusion through the oxide-fuel matrix and then through defects in cladding into the primary coolant or the spent-fuel-pool coolant. Unlike the noble gases that are continuously discharged from the primary coolant BWR, most of the ^{85}Kr is expected to remain in the primary coolant system, distributed between gaseous and liquid-solution phases as a result of pressurization until refueling or coolant letdown and purification, when the pressure is reduced or the gases are stripped out of the coolant to appear in gaseous waste streams. Since the noble gases are in solution in the primary coolant under pressure in a PWR, their appearance mechanism also includes leakage of primary coolant and vaporization into gaseous waste streams.

The amount of ^{85}Kr appearing in reactor effluents depends mainly on the cladding integrity. Since the total amount of ^{85}Kr produced by fission is 4×10^5 Ci/1000 MWe-yr, a cladding failure of 1%, as recently experienced in some PWRs (Locante 1973), would result in the discharge of about 4×10^3 Ci/1000 MWe-yr.

Information on observed amounts of noble gases released at PWRs are available in the data collected for several reactors (DRO, Logsdon 1971), but it is limited to sums of noble and activation gases (the amounts of ^{85}Kr alone are generally not available). These data, normalized to power production, are listed in Table 11. The values are much lower than for BWRs (see Table 5), thus reflecting the decay of short-lived noble gases in the sealed primary system of the PWR, which is opened only at intervals.

TABLE 11. Observed Release of Fission-Product and Activation Gases at PWRs

Plant	Gross Power, MWe	Activity, Ci/1000 MWe-yr			
		1969[a]	1970[a]	1971[b]	1972[b]
Maine Yankee	863	-	-	-	38[d]
Palisades	811	-	-	-	2,300
Yankee Rowe	185	30	37	770	230
Indian Point-1	275	2,900	-	2,400	3,100
Robert Ginna	520	-	38,000	98,000	40,000
Connecticut Yankee	600	440	1,700	6,600	1,300
Robinson	772	-	-	64	440
San Onofre-1	450	800	4,300	20,000	57,000
Point Beach-1 & 2	497[c] 497[c]	-	-	2,200	7,400
Surry-1	856	-	-	-	0.27[d]
Shippingport	150	0.002	0	-	-

[a]Normalized to 1000 MWe-yr from collected data (Logsdon 1971).

[b]Normalized to 1000 MWe-yr from power production data (Nucleonics Week) and collected emission data (DRO).

[c]Net output in MWe.

[d]Plants operated less than one year.

A more detailed study of the Yankee-Rowe reactor (Kahn 1971) identifies the release of ^{85}Kr in several specific gaseous streams, as summarized in Fig. 12. The total amount of ^{85}Kr released at the Yankee-Rowe reactor, normalized to power production, is about 16 Ci/1000 MWe-yr, a value that is much smaller than the 4×10^3 Ci/1000 MWe-yr estimated on the basis of 1% fuel failure. Releases at this reactor, however, appear to be generally low compared with those of others PWRs; Yankee-Rowe also had the lowest observed releases of total noble and activation gases (Table 11). Thus, the fission-product levels at Yankee-Rowe may be atypically low; the various isotopic activities found in primary coolant water were lower, by factors of 10^2 to 10^6, than values predicted by Yankee personnel for 1% fuel failure.

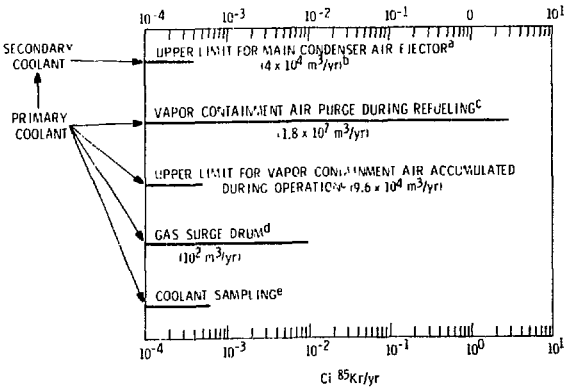

Fig. 12. Distribution of ^{85}Kr in Yankee-Rowe PWR System (Kahn 1971)

[a]Air ejector at main condenser in secondary coolant system continuously discharges noncondensible gases, including some that have leaked from primary to secondary system.

[b]Volumes are for air at STP.

[c]Vapor containment air includes radioactive gases from accumulations of primary coolant system leaks during reactor operation and releases during refueling periods. Discharge may occur four times per year.

[d]Gas surge drum collects hydrogen-bearing gases from primary coolant system and is discharged infrequently.

[e]Primary coolant sampled once-daily releases small volume of gas.

The ^{85}Kr in the reactor containment atmosphere arises from leaks of primary coolant during operation and also from release of gases during refueling. The analyses showed that $<5 \times 10^{-4}$ Ci of ^{85}Kr had accumulated from primary coolant leaks, whereas 2.8 Ci of ^{85}Kr had been released into containment air during refueling activities. The conclusions of the study of the Yankee-Rowe reactor were that the most important ^{85}Kr-bearing effluent is the air used to purge the reactor containment during refueling, resulting in a stream containing 2.8 Ci of ^{85}Kr in 1.8×10^{10} liters of air per year. However, the off-gas of the primary coolant system (gas decay tank or surge drum) has been considered to be a more likely source of ^{85}Kr (see next section).

2. Pathways of ^{85}Kr

The major pathways of ^{85}Kr in a conventionally managed PWR are diagrammed in Fig. 13. The sources considered are introduction of fission-product gases into primary coolant by diffusion through defective cladding, introduction into secondary coolant by leakage of primary coolant into secondary coolant through steam generator defects, and introduction into coolant of the spent-fuel pool by diffusion through defective cladding during spent-fuel storage.

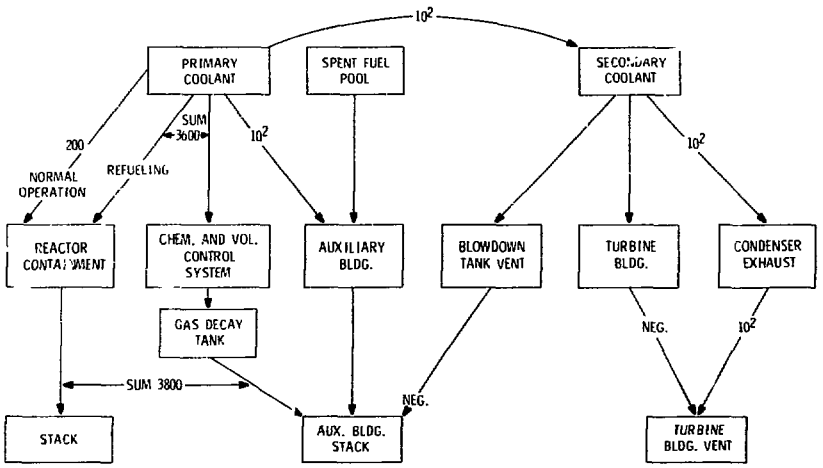

Fig. 13. Transport of ^{85}Kr in a Typical 1000 MWe PWR (amounts in Ci ^{85}Kr/1000 MWe-yr, based on assumptions outlined in Table 10; NEG = negligible amount)

Since ^{85}Kr is in solution under pressure in the primary coolant during normal operation, any path available to primary coolant during normal operation is considered to be a possible path for ^{85}Kr.

Six possible paths of coolant out of the primary system are considered: (1) continuous leakage into the auxiliary building where it would be continuously discharged into the ventilation stream; (2) continuous leakage into the reactor containment where the accumulated gas would be discharged infrequently, perhaps four times per year; (3) release to containment during shutdown, *e.g.*, refueling (during this period, perhaps totalling 30 days per year, the primary coolant remains unpressurized and exposed to the container atmosphere, which is continuously purged); (4) periodic degassing of primary coolant system during normal operation; (5) periodic degassing of boron control system (boron concentration is conventionally controlled by removal of water through evaporation or by addition of boron and water); and (6) periodic sampling of primary coolant. The small samples of coolant will release their dissolved gases in laboratory hoods and travel to the stack through building ventilation.

Only one path of ^{85}Kr out of the secondary system is considered important, *i.e.*, continuous discharge through the main condenser exhaust. The solubility of noble gases in water of the primary coolant is probably greater than in the secondary coolant system and, hence, after entering the secondary system, the dissolved noble gases would probably rapidly enter the gas phase and be swept out mainly in the condenser exhaust. Accordingly, the amounts of noble gases transported out of the secondary system by liquid paths, *e.g.*, coolant leakage and blowdown, are considered to be relatively small.

3. Composition of Waste Streams

The expected composition of ^{85}Kr-bearing streams from a 1000-MWe PWR will vary with the extent of cladding failure and with management, *e.g.*, the frequency and efficiency of degassing the primary system and the boron control system, and the frequency and volume of blowdown. The compositions of the main effluent streams, based on 1% fuel failure and a postulated set of management conditions are shown in Table 12. One of the main points of

TABLE 12. Distribution of ^{85}Kr in PWR Effluent Streams[a]

		Amounts/1000 MWe-yr		
Stream	Constituent	Ci	Liters(STP in Total Stream	Metric Tons in Total Stream
Main Condenser Exhaust	^{85}Kr	102[b,c]		
	H_2O vapor			2[d]
	Air		3×10^{8}[e]	
Containment Atmosphere Accumulated during Normal Operation	^{85}Kr	2×10^{2}[b,f]		
	H_2O vapor			6[g]
	Air		1.2×10^{7}[h]	
Auxiliary Building Ventilation	^{85}Kr	≤ 650[b,i]		
	H_2O			5×10^{5}[j]
	Air		10^{12}[k]	
Containment Purge during Refueling Shutdown	^{85}Kr	≤ 3800[e]		
	H_2O vapor			5[m]
	Air		10^{11}[n]	
Off-Gas from Primary Coolant Letdown	^{85}Kr	≤ 3800[p]		
	N_2		10^{5}[q]	

[a]Based on ∿1% fuel failure, ∿4 x 10^3 Ci ^{85}Kr/1000 MWe-yr.

[b]Based on 4 μCi ^{85}Kr/ml of primary coolant.

[c]Assuming 65 kg/day leakage of primary coolant into secordary system.

[d]Based on water saturation at 50°C of 20 scfm air flow.

[e]Based on 20 scfm air flow.

[f]Assuming 130 kg/day leakage of primary coolant into reactor containment.

[g]Based on water saturation at 25°C of 12 x 10^6 liters of air.

[h]Based on 3 x 10^6 liter volume of containment opened 4 times per year.

[i]Assuming 77 kg/day leakage of primary coolant into auxiliary building.

[j]Based on water saturation at 25° of 80,000 scfm air flow.

[k]Based on 80,000 scfm air flow.

[m]Assuming water saturation at 25°C of 80,000 scfm air purge for 30 days/yr.

[n]Based on 80,000 scfm air purge for 30 days/yr.

[p]Assuming 3800 Ci/1000 MW-yr will be distributed between containment purge and off gas from coolant letdown.

[q]Based on volume of purge tank emptied once per year.

uncertainty is the identity of the path carrying the largest portion of ^{85}Kr. The monitoring study of the Yankee-Rowe reactor (Kahn 1971) indicated that the air purge of containment during refueling was the main stream of gaseous activity but a source term model (Binford) for a PWR indicates that the off-gas from depressurization of primary coolant and the boron control system is the main carrier of ^{85}Kr, *i.e.*, chemical and volume control operations. This is a resonable hypothesis if, in the treatment of primary coolant letdown, a volume of liquid equivalent to several reactor volumes is degassed per year (DOCKET 50382-30). For frequent or efficient degassing of primary coolant letdown, the main ^{85}Kr carrier is the off-gas from the chemical- and volume-control system, whereas when this system is infrequently and inefficiently degassed the main ^{85}Kr carrier is the air purge of containment during refueling.

For this work, the distribution of ^{85}Kr in effluent streams has been estimated by assuming an average operating concentration in the primary coolant and leak rates of primary coolant to secondary system, auxiliary building, etc. The balance of the ^{85}Kr (major portion) is assumed to be distributed between the containment purge air and decay tank, which has a cover of nitrogen to exclude oxygen and dilute the hydrogen. Estimates of possible coolant activities for ^{85}Kr in a PWR with 1% failed fuel are 4.44 μCi/ml (Gallagher, Wright) and 3.44 μCi/ml (Denton). These coolant activities are consistent with values that can be obtained from the Henry's Law constant for the $Kr-H_2O$ system, the estimated gas volume in the primary coolant system, and the ideal gas law. Thus, if 4×10^3 Ci ^{85}Kr or 0.12 mol per 1000 MWe-yr are introduced into primary coolant by 1% fuel cladding failure, then the total amount of Kr, including ^{83}Kr, ^{84}Kr, ^{85}Kr, and ^{86}Kr, and neglecting shorter-lived Kr isotopes, is 1.5 mol. If the gas space in the primary system is assumed to be 2×10^8 cm^3 (from dimensions of PWR reactor vessels (NEI) and the assumption that the gas space is one third the total volume), then the partial pressure of Kr at 300°C, from the ideal gas law, is 6×10^{-3} psia. Using this partial pressure and the Henry's Law constant (Anderson) for 300°C, *i.e.*, 1.2 to 17×10^5 psia per mol fraction of Kr in H_2O, the mol fraction of Kr (including those isotopes listed above) is 0.3 to 5×10^{-8}, corresponding to ^{85}Kr concentrations of 0.5 to 7μCi/ml H_2O. These estimates are considered maximum values; operating values would be lower because of primary coolant depressurization for refueling and maintenance.

Distribution of ^{85}Kr in effluent streams, based on concentration of 4μCi/ml in the primary coolant and the indicated leak rates to the auxiliary building, secondary system, and reactor containment, are shown in Table 13.

4. Effect of Advanced Management Schemes

In contrast to conventional PWR designs in which noble gases are accumulated under pressure in the primary system and released at periodic degassings or refuelings, advanced PWR designs provide for continuous stripping of noble gases from primary coolant. The noble gases are stripped by purging the contents of the volume-control tank with hydrogen gas, and passing the resulting mixture through a recombiner to convert the hydrogen to water (Fig. 14). Thus, the resulting effluent gas stream from the primary system is mainly stable xenon, stable krypton, and ^{85}Kr, this stream is pumped into decay tanks along with a nitrogen carrier gas at 1 atm.

The volume of noble gases from 1% failed fuel over the lifetime of

TABLE 13. Comparison of Three Estimates for Distribution of ^{85}Kr in PWR Streams (approx. % of total amount generated that is released at reactor site)

Reference	From Gas Decay Tanks	Containment Purge	Secondary System	Auxiliary Bldg. Ventilation	Coolant Sampling
Wright[a]	90	1.1	4.4	4.4	-
Kahn 1971[b]	0.33	99.6	0.013	-	0.020
d	← Sum[c] 84 →		2	14	-

[a]Estimated for a Westinghouse PWR of ~1800 MWt capacity.

[b]Found by monitoring of Yankee-Rowe PWR (600 MWt).

[c]Difference between total ^{85}Kr released and amounts released to secondary system and auxiliary building. Distribution of estimated 84% of the total ^{85}Kr released at the site between gas decay tanks and containment purge is assumed to depend on frequency and efficiency of degassing primary coolant letdown during normal operation.

[d]Calculated from assumed primary coolant concentration of 4 μCi ^{85}Kr/ml and primary coolant leakages of 65 kg/day into secondary system and 77 kg/day into auxiliary building (Table 9).

the PWR (40 yr) has been estimated to be sufficiently small that the gases could be stored on the reactor site during its entire operating lifetime (Gallagher). The estimated total burden of noble gases at the decommissioning of a 1000-MWe PWR after 40 yr of operation (Table 14), would include 5.86 x 10^4 Ci ^{85}Kr (Gallagher), 10^6 liters(STP) stable xenon, 10^5liters(STP) total krypton, and an unspecified volume of nitrogen. If these totals are normalized to a power production of 1000 MWe-yr, the estimates (per 1000 MWe-yr) are about 1.5 x 10^2 Ci of ^{85}Kr, 2.5 x 10^4 liters(STP) stable xenon, and 2.7 x 10^3 liters(STP) of total krypton.

5. Conclusions

In conventional operation of PWRs, the noble gases released into the primary coolant through cladding failures accumulate under pressure in the primary system. The release of noble gases in lesser quantities can occur in several streams, owing to leakage of primary coolant, but the major portion is expected to appear in two streams: (1) the nitrogen cover gas upon degassing primary coolant in a process sidestream for adjustment of pressure, chemical content and volume; and (2) the air purge of the reactor containment atmosphere during shutdown (*e.g.*, for refueling), which sweeps out gases that have accumulated in the containment through leakage of primary coolant during operation and those that are released when primary coolant is depressurized during shutdown. The distribution of ^{85}Kr between these two streams is expected to vary with frequency and efficiency of primary-coolant degassing during normal operation. For frequent and efficient degassing of the coolant, most of the ^{85}Kr is expected to appear in the nitrogen off-gas of the chemical- volume-control system; for infrequent and inefficient degassing in normal operation,

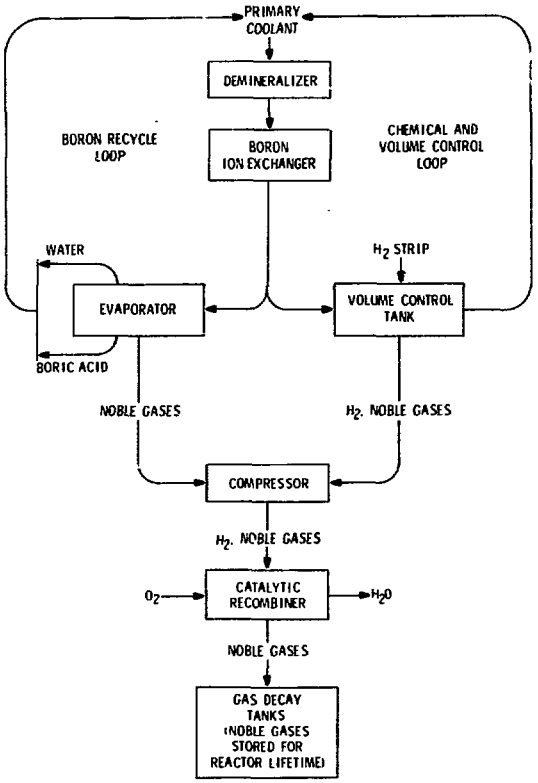

Fig. 14. Advanced PWR Noble-Gas Management Scheme

TABLE 14. Estimated Composition of ^{85}Kr-Bearing Waste of Advanced PWR

	Total Amounts		Average Burden/1000 MWe-yr	
Component	Ci	Liter(STP)	Ci	Liter(STP)
^{85}Kr[a]	5.86×10^4		1.5×10^3	
Stable Xe[b,c]	-	10^6	-	2.5×10^4
Total Kr (stable Kr + ^{85}Kr)[b,c]	-	1.1×10^5	-	2.7×10^3
N_2	-	Volume Unspecified	-	Volume Unspecified

[a]Gallagher.

[b]Cumulative after 40-yr operation, assuming decay of about two thirds of all ^{85}Kr generated (Gallagher).

[c]From fission product yields estimated for Diablo Canyon PWR (ORNL-4451).

most of the ^{85}Kr is expected to appear in the air purge of containment during refueling.

In advanced PWR concepts, noble gases released into the primary coolant will be stripped continuously from the primary coolant by a hydrogen purge. The hydrogen will then be separated from the noble gases by catalytic conversion to water, and the noble gases will be pumped, along with a carrier of nitrogen (DOCKET 50348-29, Gallagher), into decay tanks to be stored on the reactor site for the operating life of the reactor. Thus, at decommissioning of an advanced PWR, the ^{85}Kr would be present in a mixture with stable xenon, stable krypton, and the nitrogen carrier gas.

IV. BEHAVIOR OF TRITIUM AND ^{85}Kr IN LIQUID METAL FAST BREEDER REACTORS

An LMFBR of 1000-MWe capacity is not expected to start up before the early to mid-1980's (AIF); hence, information on the behavior of tritium and noble-gas fission products in a large commercial LMFBR is limited at this time to predictions. Also, because the design of a large LMFBR is not firm, these predictions are necessarily less detailed and less specific than predictions of the behavior of tritium and noble gases in light-water reactors, for which operating and monitoring data are available. In the following sections, the behavior of tritium and ^{85}Kr will be deduced from experimental observations of tritium and noble-gas behavior in smaller, sodium-cooled fast-fission facilities and also from design concepts for a 1000-MWe LMFBR recently prepared by a number of contractors (AI-AEC-127-92, BAW-1328, Buttrey, CEND-337, GEAP-4418, GEAP-5678, WARD-2000-97).

A number of recent efforts to model the behavior of tritium in an LMFBR have been described (see Section IV.C.2.); however, little information is available on the behavior of noble-gas fission products.* This discussion is not intended to anticipate or to duplicate the detailed transport calculations used in the construction of such models; it is intended, rather, to correlate the predictions of main transport paths, quantities, and compositions of tritium- and ^{85}Kr-bearing streams in a large LMFBR from available experimental data and calculations relevant to an LMFBR system.

A. Liquid Metal Fast Breeder Reactor System Descriptions

1. Core and Surroundings

A number of uncertainties presently exist in the choices of design and materials for LMFBRs; however, the first large LMFBRs introduced into a commercial utility system will probably consist of a core of UO_2-PuO_2 pellets clad in stainless steel, surrounded by axial and radial blankets of UO_2 pellets

*The transport of gaseous fission products in an LMFBR has been reviewed (Keilholtz), but with emphasis on the transport following an accident. The present discussion is concerned with transport of tritium and noble-gas fission products during normal operation.

clad in stainless steel. A number of designs have been offered (Bruzzi, O'Neill, WARD-2000-97) for vented fuel pins; these incorporate various mechanical devices, essentially pressure-relief valves, that would permit volatile fission products to escape after an interim holdup time for decay of short-lived isotopes into the primary coolant, the cover gas, or a plenum associated with the pin. Although vented fuel pins have received a great deal of consideration, they will probably not be employed in first-generation LMFBRs (WASH-1509).

2. Coolant Cycle

An example of the flow pattern of the coolant and working fluid in a large LMFBR is shown in Fig. 15. The reactor system is cooled by the transfer of heat from the core and blankets to a primary coolant of liquid sodium; from the primary to the secondary coolant, also of liquid sodium; and finally, from the secondary coolant to the working fluid, namely, water/steam. Heat transfer from the primary coolant to secondary coolant occurs in an intermediate heat exchanger and heat transfer from the secondary coolant to the working fluid occurs in an evaporator, a superheater, and a reheater.

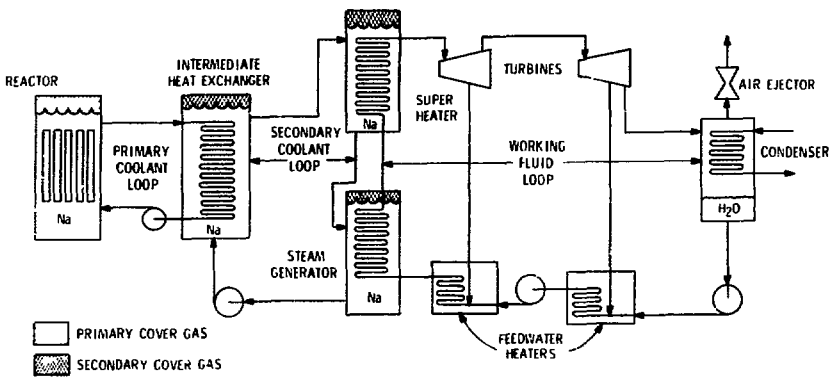

Fig. 15. LMFBR Coolant Cycles

Two arrangements for the positioning of the intermediate heat exchanger and primary coolant pump have been considered. In the pot system, the primary coolant pump and intermediate heat exchanger are both located within the reactor vessel and are thus immersed in the primary coolant. In the loop system, the pump and intermediate heat exchanger are located outside the reactor vessel; thus they conduct the primary coolant but are not immersed in it.

3. Gaseous Effluent Streams

Concepts of gaseous radwaste systems for large LMFBRs deal mainly with cover gas management, thereby reflecting the assumption that volatile fission products released from fuel will appear mainly in primary cover gas. The appearance of noble gases in secondary and steam systems would require leakage through defects in both the primary-secondary and secondary-steam in-

terfaces. The probability of leakage by this path is low, since it is mandatory that the occurrence of such defects be minimized by engineering design for reasons other than fission-product transport, *i.e.*, reactor safety related to sodium-steam reactions. In secondary and steam systems, the appearance of tritium is more probable than the appearance of noble gases because of the ability of tritium to diffuse through stainless steel.

The functions of conceptual LMFBR gaseous radwaste systems include delay by holdup in tanks or on ambient-temperature adsorption beds, filtration, monitoring, and recycle or dilution with ventilation air followed by environmental discharge. LMFBR concepts have also included the possibility of separation of xenon and krypton from argon followed by bottling of xenon and krypton for storage and recycle of argon (BAW-1328, WARD-2000-97, WASH-1509). Fig. 16 outlines proposed gaseous radwaste systems for LMFBRs.

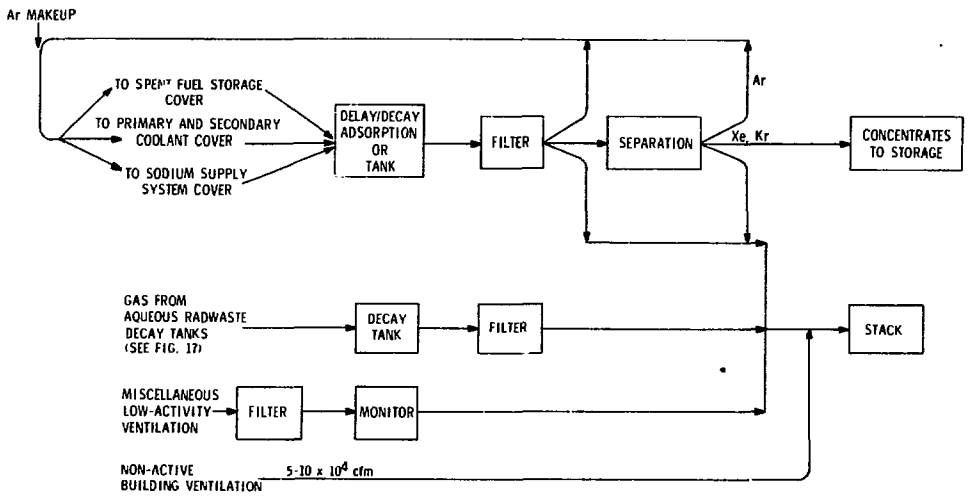

Fig. 16. Proposed Gaseous Radwaste Systems for a LMFBR

4. Liquid Effluent Streams

The functions of conceptual liquid radwaste systems for LMFBR are similar to those of light water reactor (LWR) systems; these include delay, filtration, evaporation, ion exchange, and recycle or discharge after dilution with condenser cooling water. The general level of radioactivity of aqueous LMFBR effluents is expected to be lower than for aqueous LWR effluents because the primary coolant, the initial receiver of fission products released from fuel, is remote from the aqueous system. Aqueous LMFBR streams are expected to be much less important as carriers of tritium than aqueous LWR streams. Two suggested LMFBR liquid radwaste schemes based on ion exchange (WARD-2000-97) and evaporation (BAW-1328) are outlined in Fig. 17.

B. Generation of Tritium in Liquid Metal Fast Breeder Reactors

Tritium will be generated in an LMFBR in fission of a number of actinides

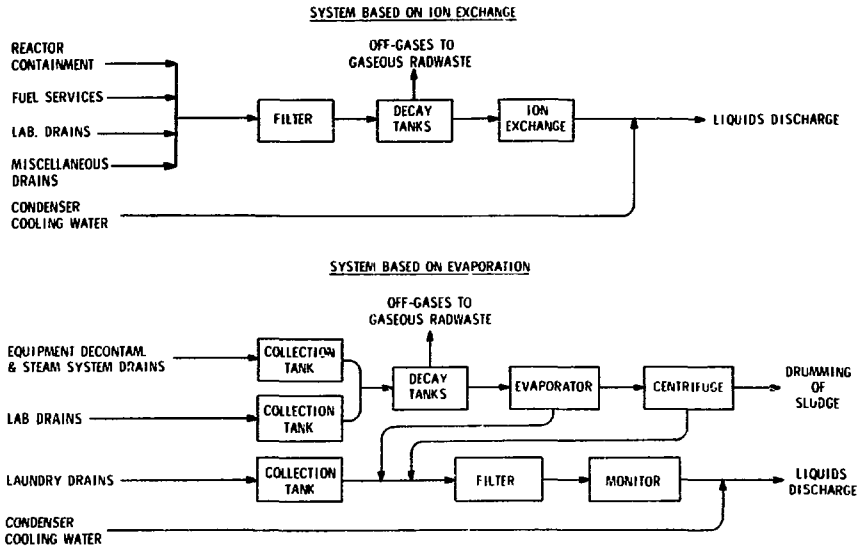

Fig. 17. Proposed Liquid Radwaste Systems for a LMFBR

and also by neutron activation of lithium and boron. Tritium generation rates can only be estimated for these processes because uncertainties exist in the fast fission yields of tritium and because information is incomplete on the neutron spectrum, and hence the activation rate, as a function of distance from the core center.

1. Fission

Most of the fission-generated tritium will originate inside the fuel and blanket rods; only a very minor fraction is expected to be generated in tramp heavy elements on the outer surface of the rods. The fast neutron fission yield for ^{239}Pu has not been experimentally determined, but upper and lower limits are estimated as follows: Tritium yields of 1.35 to 1.67 x 10^{-4} tritium atoms per fission for thermal neutron fission of ^{239}Pu have been reported (Horrocks). We assume that the lower limit for the yield of tritium in the fast fission of ^{239}Pu is an average of these values, *i.e.*, 1.5 x 10^{-4} tritium atoms per fission. We further assume that the upper limit is about 2.3 to 2.4 times the thermal yield, on the basis of the ratio of the fast and thermal tritium yields for ^{235}U (Fluss). Thus the limits of the tritium yield in fast neutron fission of ^{239}Pu are assumed to be 1.5 to 3.5 x 10^{-4} tritium atoms per fission. Since a high fraction of fissions in an LMFBR are expected to occur in ^{239}Pu, the total rate of generation by fission is estimated from these limits, and the suppositions that 2.7 x 10^{21} fissions produce 1 MWt-day of power (*i.e.*, 200 MeV/fission) and that a 1000-MWe LMFBR is rated at 2500 MWt, to be 2 x 10^{4} to 4 x 10^{4} Ci tritium per 1000 MWe-yr.

2. Boron Activation

The total generation rate of tritium in an LMFBR will be greatly affected by the choice between tantalum and boron carbide (B_4C) control rods.

No tritium is formed in the reaction of neutrons with tantalum, $^{181}Ta(n,\gamma)^{182}Ta$. On the other hand, the absorption of neutrons by B_4C produces tritium by the reaction $^{10}B(n,2\alpha)^3H$. Boron carbide control rods have been specified for the AI Reference Oxide Reactor (Buttrey), and also for the Fast Flux Test Facility (HEDL-TME-71-46), and tantalum control rods have been specified in the Follow-On concepts (AI-AEC-12792, BAW-1328, CEND-337, WARD-2000-97). Some of the concepts, however, include tantalum rods for burnup control and B_4C rods for scram or safety control. The amount of tritium produced in B_4C rods used only for scram is difficult to predict since the exposure will be unpredictable, but the exposure, and hence the rate of tritium production, should be small. Although relatively large quantities of tritium are expected to be formed in B_4C control rods, recent experimental results indicate that tritium will probably not be released from the boron carbide matrix (Ebersole). The relatively smaller amounts of tritium formed by neutron activation of boron impurities in fuel and in primary coolant, however, are expected to appear in reactor effluents.

Estimation of the tritium production rates in B_4C control rods is approximate because of incomplete information on radial dependence of flux and neutron spectrum, and on the quantities of boron involved. One estimate can be obtained on the basis of an assumed ratio of tritium produced from boron activation to tritium produced from fission, indicative of the balance between control and fission. The amounts of tritium produced by fission and boron activation have been estimated for two fast-neutron facilities, EBR-II and FFTF (Sehgal). The ratio for these two facilities are similar, about 2.4 for FFTF (300 MWt) and about 2.7 for EBR-II (62.5 MWt); these values are subject to uncertainties in the fast fission yield of tritium, the cross section for the boron activation reaction, and the flux above threshold for the reaction. If the ratio can be assumed to be similar for a 1000-MWe LMFBR, then the production of tritium in B_4C control rods would be $\sim$65,000 Ci/yr ($\sim 2.6 \times \sim 2.5 \times 10^4$ Ci/yr = 6.5×10^4 Ci/yr).

Another estimate can be obtained from an assumed tritium yield per weight of ^{10}B in fast neutron flux and assumptions of the number of control rods and weight of boron in each rod. It has been estimated (Cafasso) that approximately 1300 Ci of tritium would be produced by irradiation of 1 kg of ^{10}B in a fast flux of 10^{15} n cm^{-2} sec^{-1} for 6 months. If a control rod for a large LMFBR contains 1.3 kg. of ^{10}B, as in calculations for FFTF(Sehgal), and if a large LMFBR has 19 control rods, as specified for the AI Reference Oxide Reactor (Buttrey), than the amount of tritium formed in control rods would be 64,000 Ci/yr, which is in the range of the first estimate.

The tritium generated from neutron activation of the boron impurity in the sodium coolant, which is a small fraction of the total tritium generated in an LMFBR, can be estimated from design values for conceptual reactors. For the calculation, it is assumed that the concentration of boron in sodium is 25 ppm, a RDT purchase specification (RDT, 1973). The concentration in an operating LMFBR will probably be lower; the concentration in SEFOR was reported to be 2 ppm (Kunkel), and the concentration in EBR-II was reported to be <0.05ppm (Olson 1972). Also, for this calculation, the neutron flux in sodium is taken from a concept of a 1000-MWe LMFBR (GEAP-5618) that includes estimates of the neutron flux. The cross section for the reaction $^{10}B(n,2\alpha)^3H$ is assumed to be 30 mb., similar to values used in other recent calculations of tritium generation in fast fluxes (Kunkel, Sehgal). If the average fast flux above 1 MeV in the coolant is assumed to be 10^{13} n cm^{-2} sec^{-1} and the volume of sodium in the

primary coolant is 5.5×10^4 ft^3 (GEAP-5618), then the production rate is 70 Ci/yr.

The amount of tritium formed by the same mechanism in the secondary coolant should be even smaller since the sodium inventory (1.2×10^4 ft^3) (GEAP-5618) and the neutron flux will be smaller. The amounts formed in both primary and secondary systems of a loop reactor, involving lower sodium inventories and neutron exposure times, will also be smaller than the amount calculated above for tritium produced in the primary coolant of a pot system.

Some tritium will be generated as a result of neutron activation of the boron impurity in core and blanket material. The generation rate is estimated for the AI Reference Oxide Reactor from estimates of neutron flux and actinide composition of core and blankets (ORNL-4436) and from an assumption of a maximum boron concentration of 10 ppm, a RDT specification for FFTF fuel (RDT 1971, RDT 1971A). The results of the calculations are 370 Ci/yr for the core, 200 Ci/yr for the radial blanket, and 40 Ci/yr for the axial blanket. Thus, a total of about 600 Ci of tritium per year might be generated in the core and blankets from neutron activation of the boron impurity.

3. Neutron Activation of Impurity Lithium in Coolant and Fuel

The rate of tritium generation in a 1000-MWe LMFBR by neutron activation of the lithium impurity in the primary coolant is also likely to be small. For a pot reactor with 5.5×10^4 ft^3 of liquid sodium and with the assumptions (1) that the lithium impurity is 5 ppm (RDT, 1973), (2) that for neutrons with energies below 1 MeV the average cross section is 900 mb (Stehn) and the neutron flux is 4×10^{12} n cm^{-2} sec^{-1}, and (3) that for neutrons with energies above 1 MeV the average cross section is 100 mb (Stehn) and the neutron flux is 1×10^{13} n cm^{-2}sec^{-1}, the rate of tritium generation from lithium is about 85 Ci/yr. As in the case of boron activation in the coolants, the tritium generation rate through lithium activation in the secondary coolant will be less than in the primary coolant. Also, the generation rates in both primary and secondary coolants of a loop reactor will be lower as a result of lower sodium inventories and lower neutron exposures.

The rate of tritium generation by neutron activation of lithium impurity in core and blanket materials is estimated from flux and material amounts assumed for the core and blanket of the AI Reference Oxide Reactor (ORNL-4436) together with the assumed maximum concentration of 10 ppm Li in oxides (RDT 1971), RDT 1971A). The results are 720 Ci/yr for the core, 390 Ci/yr for the radial blanket, and 290 Ci/yr for the axial blanket. Thus, the total is about 1400 Ci/yr of tritium formed from neutron activation of lithium impurity in the core and blankets.

The estimated values for tritium generation rates in a 1000-MWe LMFBR are summarized in Table 15.

C. Appearance of Tritium in Liquid Metal Fast Breeder Reactor Waste Streams

The appearance of tritium in the LMFBR primary coolant, which is the initial step in transport and distribution in the system, occurs by escape of fission-product tritium from fuel rods and by neutron activation of boron and impurities in contact with the coolant. The escape of fission-product tritium from fuel rods can occur through cladding defects, but more importantly, by

TABLE 15. Estimated Rate of Tritium Generation and Rate of Appearance in Waste Streams of LMFBRs

Source	Expected Rate of Generation, Ci/1000 MWe-yr
Fission	$2 - 4 \times 10^4$
Activation of B_4C control rods	65,000[a]
Activation of B impurity in primary coolant	≤70
Activation of B impurity in core & blanket fuel	600
Activation of lithium in primary coolant	≤85
Activation of lithium in core and blanket fuel	≤1400

[a]This tritium is expected to be retained in the control rods and not to appear in reactor effluents.

permeation of the stainless steel cladding by elemental tritium. The latter mechanism is expected to result in release of nearly all the fission-product tritium into the primary coolant. This conclusion is supported by information on the permeability of stainless steel by tritium (Chaney, Elleman), by analyses of tritium distribution in sodium-cooled reactor facilities such as EBR-II (Ebersole 1972) and SEFOR (Kunkel), and by analyses for tritium in irradiated fuel (Goode, Wozadlo). Several models for the behavior of tritium in LMFBR have been developed (Court, Drawford, Kabele, Kumar, Taylor). Some of these will be discussed in Section 2 below.

1. Observations of Tritium Transport in Related Systems

Analyses for tritium at various locations in the EBR-II system have been expressed as percentages of the total calculated to have been produced in fission (Ebersole 1971); these values are given in Table 16. These percentages, however, were based on values for thermal fission yields of tritium in ^{235}U, and are, therefore, subject to correction (Ebersole 1972) since the fission yields are now known to be greater (Dudey, Fluss). If the tritium yield for fast fission is a factor of two greater than for thermal fission, then the percentages will be about half of those listed in Fig. 9. As a result, the tritium accounted for will be a factor of two lower, and the uncertainty in tritium distribution will be greater. If the results on tritium distribution in the EBR-II complex are indicative of distribution to be expected in a 1000-MWe LMFBR, they suggest that a high percentage of the tritium formed in the fuel will pass into the primary coolant, that a large fraction of tritium in the primary coolant will be removed by the primary cold trap, that a small fraction will migrate through the primary containment to appear in the secondary coolant system, and that an even smaller fraction will penetrate the secondary containment to appear in the water/steam system.

It should be noted, however, that the distribution of tritium may depend on the type of fuel material and the reactor operating temperature. EBR-II was operated on metallic fuel at the time of the observations described above, whereas an LMFBR is expected to contain oxide or carbide fuels and to operate at higher temperatures than EBR-II.

TABLE 16. Distribution of Tritium in EBR-II[a]

Location	Percent of Fission-Generated Tritium[b]
Fuel	20-25
Primary Cold Trap	68
Primary Sodium	4
Secondary Sodium	0.2
Water/Steam System	0.5
Primary Cover Gas	10^{-4}
Secondary Cover Gas	10^{-5}
Shield Cooling Air	1

[a] Ebersole 1972).

[b] On the basis of tritium yield in thermal fission of ^{235}U. Since yields of tritium in fast fission are greater (Dudey, Fluss), these values should be lower (Ebersole 1972).

The amounts of tritium found in primary sodium of the SEFOR facility were almost equal to the total amounts of tritium calculated to have been formed in fission, suggesting almost quantitative release of tritium from fuel into the sodium. These calculations, however, were also based on the yield of tritium for thermal fission (Kunkel), and are therefore subject to correction.

Analyses of tritium remaining in irradiated fuels agree with the conclusion, from analyses of coolants, that a high percentage of tritium escapes from fuel clad with stainless steel. Stainless steel-clad oxide fuels, irradiated in a fast flux to burnups of 46,000 to 79,000 MWd/metric ton at linear powers of 11 to 14 kW/ft, have been observed to contain 1% of the calculated amounts of fission-product tritium (Wozadlo). Other analyses of LMFBR-type fuels, irradiated at powers of 9 to 16 kW/ft, indicated that less than 5% of the theoretical amounts of tritium expected from fission were present (Goode).

The expected distribution of tritium in LMFBR cover gas and coolant can also be estimated from their volumes and the stability of sodium hydride. Measurements by several investigators of the dissociation pressure of NaH in equilibrium with liquid sodium have been used to estimate the dissociation pressure of NaT in equilibrium with sodium (Wozadlo). These observations combined with the assumption that the cold-trap temperature in an LMFBR would be 121°C were used to estimate that the partial pressure of tritium in primary cover gas would be 0.5 to 1.0 x 10^{-3} Torr for steady-state operation (Wozadlo). This partial pressure of tritium in the cover gas of a 1000-MWe LMFBR, together with an assumption that the volume of primary cover gas* would be 1.4 x 10^8 cm^3, indicates that the steady-state inventory of tritium in primary cover gas would be 200 Ci, but the amount can be expected to vary with cover-gas processing frequency.

*One 1000 MWe LMFBR concept specifies a volume of 1.9 x 10^8 cm^3 (O'Neill); another concept specifies a volume of 7.1 x 10^8 cm^3 (BAW-1328).

The estimated partial pressure of tritium in cover gas cited above can also be used, in conjunction with Sieverts' law, $S = K_p^{1/2}$,* to estimate the corresponding inventory of tritium in primary coolant, assuming that the steady-state concentration of tritium is its solubility at the temperature of the primary cold trap, which is expected to be a point of accumulation of tritium. In the absence of a Sieverts' law constant for tritium, the value for hydrogen, K = 5 ppm (weight)-Torr$^{-1/2}$ (Vissers) was used to estimate a tritium concentration of 0.1 g or 10^3 Ci/10^6 g Na. If the primary coolant volume of 1000-MWe LMFBR is 6.8 x 10^8 cm^3 (Taylor), then the primary-coolant inventory of tritium would be 6 x 10^5 Ci. This amount, however, corresponds to about 30 times the annual amount expected to be introduced into the primary coolant of a 1000-Mwe LMFBR. This calculation thus indicates that in the absence of other factors, the appearance of tritium in the cold trap and the attainment of the steady-state partial pressure of tritium in the cover gas, cited above, would require the accumulation of all tritium released in 30 years of operation. The concentration of hydrogen in the LMFBR primary system, however, is expected to affect the behavior of tritium (*e.g.*, coprecipitation of tritium with NaH in the cold trap), but neither the concentration nor its effects are clearly defined at this time. The estimates cited above for concentrations and partial pressures of tritium in an LMFBR, owing to their equilibrium basis, *i.e.*, the dissociation pressure of NaT, are probably lower limits. In actual practice, at any given time, the concentration of tritium in sodium and partial pressure of tritium in cover gas will probably depend on the balance between the rate of diffusion out of fuel pins, the cover gas processing rate, and the rate of deposition in the cold trap, which is, in turn, a function of cold trap throughput rates and efficiencies. The latter philosphy was used to estimate the appearance of tritium in the steam system of an LMFBR as a function of throughput rates of primary and secondary cold traps (Taylor).

2. Models of Tritium Transport in Related Systems

Models of the tritium behavior in the Fast Flux Test Facility (FFTF) that consider the release of tritium by diffusion through sodium-carrying pipes in both the primary and secondary coolant loops have been described. Since the FFTF is not designed to produce electric power, it has no water/steam system. Heat is transported from the primary coolant to the secondary coolant through the intermediate heat exchanger (IHX), and heat is transported from the secondary coolant to the environment through a dump heat exchanger (DHX). One model (Court) showed that the sum of tritium collected in primary and secondary cold traps would be about 94% of the total introduced into the primary coolant, with about 3% escaping to the atmosphere through pipes carrying primary and secondary sodium, and about 3% escaping to the atmosphere from the DHX. Presumably this latter amount would appear in the water/steam system of a similar system designed to produce electric power.

Another model of FFTF (Kabele) that included the dependence of tritium distribution on cold-trapping rate and cold-trap efficiency indicated that the amounts of tritium in sodium, in gas surrounding the sodium-carrying pipes, and in releases through the DHX could account for <5% of the tritium introduced into

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*S = the concentration in sodium, K is the Sieverts' constant, and p is the equilibrium pressure in the gas phase.

the coolant, implying that the remainder, >95% of the tritium introduced to the primary system, would deposit in the cold traps.

A model of tritium behavior in a 1000-MWe LMFBR (Taylor) described the escape of tritium into the water/steam system as a function of the purification rate of primary and secondary coolant. The percentages of tritium calculated to be released from the steam system varied from about 98% for no cold-trapping to 0.7% for a cold trapping rate of 380 liters/min for both the primary and secondary systems.

A model aimed at a general description of the transport of tritium in liquid-sodium-cooled nuclear facilities has been under development at Argonne National Laboratory (Kumar). The transport paths considered in this model include core-to-primary-to-secondary-to-steam, diffusion through walls of pipes carrying both sodium and steam, loss by cover-gas purging, loss through water discharge, and deposition as a sodium compound in primary and secondary cold traps. This model assumes that the presence of hydrogen plays an important part in determining the behavior of tritium in the system, especially in processes involving deposition in the cold trap such as coprecipitation and isotopic exchange. Applications of this model to describe the behavior of tritium in EBR-II have shown that a high percentage (~95%) is expected to remain in the primary sodium or cold trap, but the effect of hydrogen content on the system has yet to be completely defined.

3. Expected Behavior of Tritium

Based on experimental observations and calculations (see preceding subsections 1 and 2), it is expected that nearly all the tritium formed in an LMFBR, except that formed by neutron activation of boron in control rods (when used), will be released into the primary coolant. The transport and distribution of tritium throughout the LMFBR system is expected to result in an inventory of tritium in sodium coolants and in the cover gas. The inventory and concentration in the cover gas is expected to be relatively small and may vary cyclically with cover-gas processing operations. Suggested processing frequencies have varied from continuous processing to processing only at refueling, perhaps semiannually. The major amount of tritium is expected to accumulate continuously in the primary cold trap. If the species collected is assumed to be sodium tritide, then the weights might be several tens of grams (~10^4-10^5 Ci/1000 MWe-yr) of NaT in the primary cold trap and several grams (10^4 Ci/1000 MWe-yr) of NaT in the secondary cold trap. The contents of the cold traps may also include several kilograms of Na_2O, the amount depending on the integrity of the cover gas seal and the resulting air leak. Lesser amounts of carbon, hydrogen, and nitrogen compounds may be present in the cold traps based on the analysis of a FERMI-I cold trap (Bradley). The cold traps will probably also contain sodium and its activation products, activated corrosion products, fission products (WARD-2000-97), and a number of metallic and nonmetallic impurities in minor concentrations, based on analyses of EBR-II sodium (Olson). Small percentages of total tritium are also expected to be transported through the system by a number of possible paths, as indicated by Fig. 18. The compositions of the major tritium-bearing streams are summarized in Table 17.

D. Generation of ^{85}Kr in Liquid Metal Fast Breeder Reactors

The only site of generation of significant quantities of ^{85}Kr in LMFBRs will be in the fuel matrix, mainly as result of fission of ^{239}Pu; several

percent of the total fissions will also occur in ^{240}Pu, ^{241}Pu, ^{242}Pu, and in ^{238}U, ^{235}U, and ^{236}U (GEAP-5618). Estimates of the expected total quantity of ^{85}Kr, which will vary with the contributions from the various isotopes listed above and with design and management of core and blankets, are in the range from 2 to 3 x 10^5 Ci ^{85}Kr/1000 MWe-yr (ORNL-4436, WASH-1509). The amounts generated in LMFBRs per 1000 MWe-yr are smaller than those generated in LWRs because of the lower yield in ^{239}Pu fission and the higher thermal-to-electrical conversion efficiency in LMFBRs.

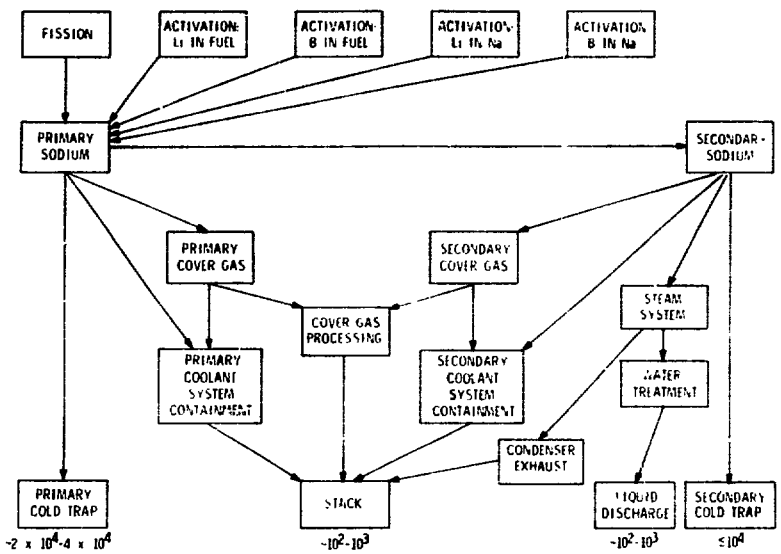

Fig. 18. Transport of Tritium in a LMFBR (amounts in Ci ^3H/1000 MWe-yr after saturation of primary sodium; coolant cycles shown in Fig. 15)

E. Appearance of ^{85}Kr in Liquid Metal Fast Breeder Reactor Streams

1. Mechanisms and Amounts

The mechanism of appearance of ^{85}Kr in LMFBR waste streams involves a chain of events starting with the fission process in the fuel matrix, migration of the ^{85}Kr atoms through the fuel matrix to the cladding, escape through cladding defects into the primary coolant, and migration as gas bubbles from the primary coolant to the primary cover gas. Perhaps the area of greatest uncertainty in this mechanism is the extent of release of fission-product noble-gas atoms from the fuel matrix. Observed releases of noble gases from irradiated oxide fuel have varied from a few percent to nearly 100% of the amounts generated (Evans, Steele, Yuill).

The percentage of failed fuel rather than the percent release from fuel matrix, mentioned above, is expected to be the determining factor in the

TABLE 17. Estimated Composition of Major Tritium-Bearing Streams of LMFBRs

Stream	Constituent	Amounts/1000 MWe-yr	
		Ci ^{3}H	Liters(STP)
Primary Cold Trap	NaT	2-4 x 10^4	-
	Na[a]	-	-
	Na_2O[a]	-	-
	NaH[a]	-	-
Primary Cover Gas	Ar	-	10^4
	T_2-HT	2-4 x 10^2	-
Primary Coolant	Na	-	10^6[b]
	NaT	6 x 10^5[c]	-
Primary Containment Ventilation	Air[a]	-	-
	T_2-HT	10^3[d]	-

[a]These materials are expected to be present in amounts which are significant but which cannot be estimated at the present time.

[b]Assumed steady-state inventory (Taylor).

[c]Steady-state inventory of primary coolant of a 1000-MWe LMFBR, based on dissociation pressure of NaT and neglecting effects of hydrogen. This inventory would not be reached until after more than 15 yr of operation (see test, Section III.C.1).

[d]From permeation of tritium through pipes carrying primary coolant, based on assumption that 1-10% of tritium introduced into primary coolant will pass through pipe walls.

release of ^{85}Kr from nonvented fuel. Thus, for a total generation rate of 2-3 x 10^5 Ci ^{85}Kr/1000 MWe-yr, the expected release of ^{85}Kr on an arbitrarily assumed basis of 1% fuel failure will be in the range from 2 to 3 x 10^3 Ci ^{85}Kr/1000 MWe-yr.

2. Pathways of ^{85}Kr

Because the design of an LMFBR of 1000-MWe capacity is subject to a number of options, the pathways of ^{85}Kr in the system are subject to some uncertainty. The design suggestions for a 1000-MWe LMFBR and associated gaseous radwaste systems (BAW-1328, WARD-2000-97) have been used to construct the diagram of possible pathways of ^{85}Kr shown in Fig. 19.

3. Composition of Main ^{85}Kr-Bearing Stream

Control of ^{85}Kr in the LMFBR will center around cover-gas management. Various suggestions for the frequency of processing the cover gas have included continuous processing (BAW-1328), or purge, and periodic processing performed at refueling (O'Neill). Suggested operations for cover gas cleanup have included the following:

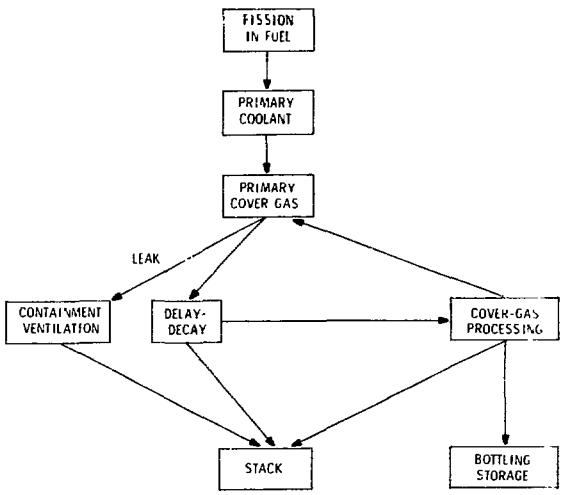

Fig. 19. Transport of ^{85}Kr in an LMFBR System

(1) Passage through ambient-temperature charcoal beds (WARD-2000-97), apparently providing only holdup and decay of short-lived isotopes, followed by dilution with air and discharge to atmosphere.

(2) Diffusion through a bank of permselective membranes, resulting in concentration of ^{85}Kr in a smaller volume of cover gas, which can then be bottled and sent to storage (BAW-1328).

(3) A combination of operations, consisting of adsorption on ambient-temperature charcoal beds for delay and decay of short-lived isotopes followed by cryogenic distillation of argon, resulting in a concentration of ^{85}Kr in a smaller volume of gas, which can be bottled and sent to storage. This latter combination has been suggested for the treatment of cover gas in the FFTF (Foley) and also in the LMFBR demonstration plant (WASH-1509).

For stable isotopes and 10.76-yr ^{85}Kr, the generation rates multiplied by a release fraction are a good measure of the amounts that require long-term storage or disposal operations. For shorter-lived isotopes, on the other hand, the amounts that will become involved in collection and disposal operations will be much less than those generated. Moreover, some of the shortest-lived isotopes are generated in large amounts, but the amounts encountered in separation and disposal operations will be negligibly small owing to decay in transit. The in-transit decay time will greatly affect not only the total amounts of noble-gas isotopes encountered at the collection or disposal point, but also, because of the variance in half-lives, the composition of the mixture.

Holdup time in leaking, nonvented fuel pins has been estimated to be 10 min (O'Neill, WARD-2000-97). The average time for disengagement of noble gases from the sodium in SEFOR was found by experiment to be about 5 min (Regimbal). Although the disengagement time is expected to be longer for the larger sodium pool of a 1000-MWe LMFBR, estimates for the deentrainment of noble

gases (identity not defined) from primary coolant in the 1000-MWe concept of GE assume that a maximum of only 2×10^{-4}% of gas released is entrained at any time in the coolant (O'Neill). Since the solubility of ^{85}Kr in sodium is small, (Vogel), the deentrainment is thought to consist largely of mechanical separation (Sowa).

Conceptual studies of the 1000-MWe LMFBR have included estimates of equilibrium activities in cover gas for defective fuel (assuming 10-min holdup in the pin) and for vented fuel (assuming 5-day holdup in the pin). Examples are given in Table 18, and volumes of fission-product isotopes, corresponding to the activities, have been added. These calculations are based on a GE model of fission gas release (O'Neill), wherein the fraction of a gaseous radioactive isotope released from fuel has been expressed as a function of the release fraction of a stable isotope and the half-life of the radioactive isotope.* For a stable-isotope release fraction of 0.35, 0.15 MCi/yr or 10^5 cm^3 (STP)/yr of ^{85}Kr and 55.2 MCi/yr or 5×10^4 cm^3 (STP)/yr of ^{133}Xe will be collected if the cover gas of a reactor using vented fuel were processed at six-month intervals. A Westinghouse LMFBR concept (WARD-2000-97) offers a comparison with these figures; it is based on a release fraction of 0.5 for stable isotopes and include predictions that can be used to calculate that 0.169 MCi/yr or 1.14×10^5 cm^3 (STP) of ^{85}Kr and 37.6 MCi/yr or 3.4×10^4 cm^3 (STP) of ^{133}Xe will be released using vented fuel.

The radioactive noble gases collected for storage will be accompanied by relatively larger volumes of stable noble-gas fission products. The volumes of stable isotopes produced in a 1000-MWe LMFBR have been approximated from tabulated fission-product yields for the AI Reference Oxide Reactor at a load factor of 0.85 (ORNL-4436). The amounts generated have been multiplied by factors of 0.01, corresponding to 1% leakage and 0.35, corresponding to 35% release from matrix, to make them comparable with the treatment of radioactive isotopes tabulated for release at the GE reactor (Table 18). The volumes of stable isotopes are shown in Table 19. Also, the volumes of gases that would require long-term storage are summarized in Table 20.

If noble gases are collected at the reactor site for long term storage, it would seem advantageous to provide some decay before final packaging. Table 21 shows an example, *i.e.*, the noble gas fission products that would be present in cover gas that had been removed from a reactor and allowed to decay 30 days before processing. The effect of 30 days cooling is evident in the summaries of Table 22. Although the total yield of radioactive xenon isotopes is larger than that for ^{85}Kr, the xenon isotopes are reduced by these decay times to amounts that are small compared with the amount of ^{85}Kr.

4. Conclusions

A total of 0.2-0.3 MCi/yr of ^{85}Kr will be generated in a 1000-MWe LMFBR; some will be released into reactor cover gas and the reminder will appear at the reprocessing plant. Less than 1% of that formed should appear in the cover gas through leakage from defective, nonvented fuel, but vented-fuel-pin designs have been assumed to release (for stable and long-lived isotopes)

*$F = F^\circ e^{-\lambda t}$ = release fraction of a given radioactive isotope, where F° = release fraction of stable isotope, λ = decay constant, and t = apparent holdup time.

TABLE 18. Radioactive Noble-Gas Fission Products in Cover Gas of a 1000-MWe LMFBR[a]

Isotope	Half-Life	MCi/yr	cm^3(STP)/yr	Moles/Mole Cover Gas[c]
		Leaking[b] Pins		
^{89}Kr	3.2 m	0.232	0.0880	2.28×10^{-10}
^{137}Xe	3.8 m	0.144	0.0668	1.73×10^{-10}
^{135m}Xe	15 m	0.177	0.0322	8.34×10^{-11}
^{138}Xe	17 m	0.564	1.14	2.95×10^{-9}
^{87}Kr	1.3 h	0.382	3.56	9.22×10^{-9}
^{83m}Kr	1.86 h	0.069	0.918	2.38×10^{-9}
^{88}Kr	2.8 h	0.546	10.9	2.82×10^{-8}
^{85m}Kr	4.4 h	0.225	7.24	1.88×10^{-8}
^{135}Xe	9.2 h	0.950	62.0	1.61×10^{-9}
^{133m}Xe	2.3 d	0.0245	9.87	2.56×10^{-8}
^{133}Xe	5.3 d	0.996	902	2.34×10^{-6}
^{131m}Xe	12 d	0.003	6.36	1.65×10^{-8}
^{85}Kr	10.7 y	0.0015	1000	2.59×10^{-6}
		Vented[d] Pins		
^{135}Xe	9.2 h	0.0105	0.684	1.77×10^{-9}
^{133m}Xe	2.3 d	0.572	230	5.96×10^{-7}
^{133}Xe	5.3 d	55.2[e]	50,000[e]	1.29×10^{-4}
^{131m}Xe	12 d	0.230	474	1.23×10^{-6}
^{85}Kr	10.7 y	0.150[f]	100,000[f]	2.59×10^{-6}

[a]Calculated for values listed for GE concept O'Neill) based on semiannual processing of accumulated activities in non-purged cover gas.

[b]Assuming release fraction of 0.35 for stable isotopes and 1% defective fuel.

[c]Based on cover gas volume of 6,830 ft^3 of argon with twice-a-year replacement.

[d]Assuming release fraction of 0.35 for stable isotopes.

[e]Predictions for ^{133}Xe release, assuming release fraction of 0.50 for stable isotopes from vented fuel, are 37.6 MCi/yr and 3.4×10^4 cm^3 (STP)/yr; calculated from values listed in WARD-2000-97.

[f]Predictions for ^{85}Kr release assuming release fraction of 0.50 for stable isotopes from vented fuel, are 0.169 MCi/yr and 1.14×10^5 cm^3 (STP)/yr, calculated from predicted yields in WARD-2000-97.

TABLE 19. Stable Noble-Gas Fission Products in Cover Gas of a 1000-MWe LMFBR[a]

Isotope	Leaking Pins[b,c]	
	Liters (STP)/yr	Moles/Mole Cover Gas[d]
^{83}Kr	1.45	3.76×10^{-6}
^{84}Kr	2.50	6.48×10^{-6}
^{86}Kr	2.30	5.96×10^{-6}
^{130}Xe	0.13	3.37×10^{-7}
^{131}Xe	9.34	2.42×10^{-5}
^{132}Xe	13.50	3.50×10^{-5}
^{134}Xe	18.90	4.90×10^{-5}
^{136}Xe	16.50	4.28×10^{-5}

[a]Calculated from weights of fission products at discharge for AI Reference Oxide Reactor at 85% load factor (ORNL-4436).

[b]Assuming 1% defective fuel and release fraction of 0.35.

[c]Corresponding values for vented pins, assuming release fraction of 0.35 for stable isotopes, are larger by a factor of 10^2.

[d]Based on a cover gas volume of 6,830 ft^3 of argon for GE pot-type concept (O'Neill) with processing and complete replacement of cover gas twice a year.

TABLE 20. Volumes of Noble Gases Requiring Long-Term Storage from Collection at a 1000-MWe LMFBR

	Leaking Pins[a,b]	
	Liters (STP)/yr	Moles/Mole Cover Gas[c]
^{85}Kr[d]	1	2.59×10^{-6}
Stable Isotopes[e]	64.6	1.67×10^{-4}
Sum[f]	65.6	1.70×10^{-4}

[a]Assumes release fraction of stable isotopes = 0.35 with 1% defective fuel; actual operation should involve a smaller percentage of defects.

[b]Corresponding values for vented pins, assuming release fraction of 0.35 for stable isotopes, are larger by a factor of 10^2.

[c]Assumes cover gas is 6,830 ft^3 of argon (O'Neill). Volume of cover gas in loop concept is expected to be smaller.

[d]Calculated from yields predicted for the GE conceptual reactor (O'Neill).

[e]Calculated from weights of fission products tabulated for the AI Reference Oxide reactor (ORNL-4436).

[f]Sums may be greater than the values listed since some cover gas, depending on the collection process and its efficiency, will accompany the noble gas fission products.

TABLE 21. Major Radioactive Noble-Gas Fission Products in Cover Gas Based on Six-Month Operation and 30-Day Cooling[a]

Radioactive Isotope	Defective Fuel		
	MCi/yr	Moles/yr	Moles/Mole Cover Gas[b]
^{133m}Xe	3.6×10^{-6}	6.2×10^{-8}	3.6×10^{-12}
^{133}Xe	19.2×10^{-3}	7.8×10^{-4}	4.5×10^{-8}
^{131m}Xe	5.4×10^{-4}	5.0×10^{-5}	2.9×10^{-9}
^{85}Kr	15.0×10^{-4}	4.4×10^{-2}	2.6×10^{-9}
Stable (Total)[c]		2.88	
	Vented Fuel		
^{133m}Xe	8.2×10^{-5}	14.8×10^{-7}	8.6×10^{-11}
^{133}Xe	10.6×10^{-1}	4.4×10^{-2}	2.6×10^{-6}
^{131m}Xe	4.0×10^{-2}	3.8×10^{-3}	2.2×10^{-7}
^{85}Kr	15×10^{-2}	4.4	2.6×10^{-4}
Stable (Total)[c]		144	

[a]Assuming cover gas management involves removal of accumulated gases from 1000-MWe GE conceptual reactor (O'Neill) after 6 months of non-purge operation, then cooling 30 days before processing for final storage. Effect of 30-day cooling can be assessed by comparison with values in Table 18.

[b]Argon in most concepts. Volume = 6,830 ft^3 for GE conceptual reactor (O'Neill).

[c]Calculated from tabulated values for 1000-MWe AI Reference Oxide Reactor (ORNL-4436).

TABLE 22. Summary of Activities to be Handled in Semiannual Processing of LMFBR Cover Gas (MCi/yr)

Vented Fuel		Defective Non-Vented Fuel	
No Decay	30-day Decay	No Decay	30-day Decay
56[a]	1.2[b]	4.3[a]	0.021[b]

[a]Sums from Table 18.

[b]Sums from Table 21.

35% (O'Neill) to 50% (WARD-2000-97). Much larger quantities of other radioactive isotopes of krypton and xenon are generated, but owing to their short half-lives, many will decay before they reach a collection point. Thus, if a collection process were used at a reactor employing vented fuel, the only radioactive isotopes that would need to be considered are ^{135}Xe, ^{133m}Xe, ^{133}Xe, ^{131m}Xe, and ^{85}Kr.

Based on semiannual processing of cover gas, a nonvented-fuel concept requires that 1.5×10^3 Ci/yr or 1 liter (STP)/yr of ^{85}Kr, 9.96×10^5 Ci/yr or 0.9 liter (STP)/yr of ^{133}Xe, and 66 liters (STP) of stable xenon and krypton isotopes be collected yearly at the reactor, whereas a vented-fuel concept requires that 1.5×10^5 Ci/yr or 100 liters (STP)/yr of ^{85}Kr, 5.52×10^7 Ci/yr or 50 liters (STP)/yr of ^{133}Xe, and 6.6×10^3 liters (STP) of stable xenon and krypton isotopes be collected yearly at the reactor.

Noble gas fission products collected at the reactor would be accompanied by some reactor cover gas, argon or helium, the quantities depending on the collection process used and its efficiency.

Handling of fission-product gases collected from continuous processing of the argon cover gas involves more radioactivity. Following 30-day decay, both methods of cover gas management result in equivalent noble-gas inventories, related only to the type of fuel. Furthermore, in both types of cover gas management, vented fuel requires the handling of 100 times as much gas as non-vented fuel for which a 1% fuel failure rate is assumed.

V. BEHAVIOR OF TRITIUM AND NOBLE GASES IN HIGH-TEMPERATURE GAS-COOLED REACTORS

As in the case of LMFBRs, a high-temperature gas-cooled reactor (HTGR) of 1000-MWe capacity has yet to be built and operated. A current timetable (NEI) places the time of operation of a large HTGR in 1979; thus an appraisal of the behavior of tritium and noble gases in such a plant is limited to small-scale investigations and theoretical modeling. The reactor model used here is based on design parameters for the proposed Philadelphia Electric 1160-MWe HTGR power plant (Gulf Oil Corp. Facts). Reference will be made to the completed 330-MWe Fort St. Vrain (FSV) plant of the Public Service Company of Colorado and also to operating experience with the 40-MWe Peach Bottom I plant of the Philadelphia Electric Company.

The major difference between HTGRs and light-water and liquid-metal-cooled reactors (other than the use of helium as a coolant) is in the materials and design of fuel assemblies. The basic component of a HTGR fuel assembly is a spherical fuel particle made up of a fuel "kernel" (metal oxide or carbide) covered with layers of graphite in the "BISO" type, and with an additional layer of ceramic silicon carbide in the "TRISO" type. Because the economics of the HTGR is based on the conversion of natural thorium to ^{233}U for reuse, there will be two types of fuel used for kernels. One, called the fissile particle, will contain ^{235}U or recycled ^{233}U. The other type, a fertile particle, will contain only natural thorium. In the reference model HTGR, both fissile and fertile particles will be of the BISO variety (see Fig. 20) and the fissile fuel will be ^{233}U. A mixture of both particles will be compacted into fuel rods with a graphite matrix and inserted into graphite-block fuel assemblies. Details of assembly design are presented in a later section.

Recent preliminary experimental results (GULF-GA-A12222), presented later, indicate that significant amounts of fission-formed tritium will diffuse through intact coatings of both BISO and TRISO particles. It had been postulated earlier (DOCKET-50267-14, Goodjohn) that the only release of tritium from the fuel particles during irradiation would result from particle failure (cracking) and that the amounts would be insignificant.

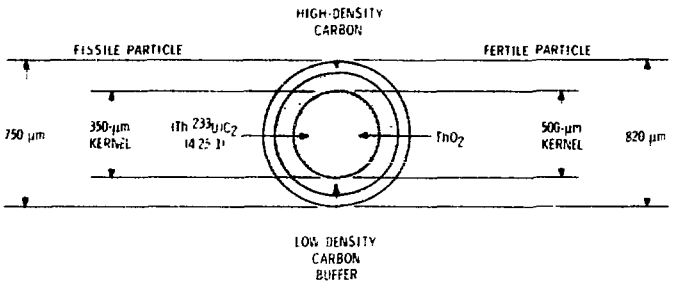

Fig. 20. BISO Fuel Particles for Proposed 1160-MWE Philadelphia Electric HTGR (Gulf Oil Corp. Facts)

Tritium can also appear in the coolant as the result of various neutron-activation reactions. The $^{6}Li(n,\alpha)^{3}H$, $^{10}B(n,2\alpha)^{3}H$, and the $^{10}B(n,\alpha)^{7}Li(n,n\alpha)^{3}H$ reactions can occur from lithium and boron impurities in the graphite of the fuel assemblies. Also, natural helium contains 1.3×10^{-4}% ^{3}He, which gives rise to the activation process $^{3}He(n,p)^{3}H$. One reference (GULF-GA-A12222) suggests that the activation of helium might be the major source of tritium in the coolant of an HTGR.

Noble gases will be generated in the HTGR only through fission. Their appearance in the helium coolant is expected to result solely from fuel-coating failure. The possibility of noble-gas diffusion through the coating is not approached in the literature. Only one reference (Goodjohn) could be found on the possible release of noble gases from HTGR fuels. In the description of a helium-coolant purification system, it was stated that a high-efficiency cryogenic charcoal bed might collect, during a year's time, 1.1×10^{4} Ci of ^{85}Kr out of a total of 5.5×10^{5} Ci generated annually in the fuel. This prediction was based on the reference design value for fuel-particle failure. Elsewhere (DOCKET-50267-15), the efficiency of the cryogenic adsorber was stated to be more than 99%, indicating that Goodjohn predicted a failure rate of about 2% for reference 1000-MWe HTGR fuel.

The helium-coolant purification system mentioned above is designed to remove not only ^{85}Kr, but also tritium and other contaminants from the primary coolant of HTGRs. The cleanup system is part of the Peach Bottom I and FSV reactor plants and will be incorporated into the design of future plants (DOCKET 50267-44, Goodjohn, Gulf Oil Corp. Facts). Management practice includes a 60-day retention of the collected (six-month collection) noble gases prior to controlled release and permanent storage of the chemically bound tritium.

A. High Temperature Gas Cooled Reactor System Descriptions

1. Core and Surroundings

The core of a reference 1160-MWe HTGR, which is enclosed in a prestressed concrete reactor vessel (PCRV), contains hexagonal blocks of graphite 31.2 in. high and 14.2 in. across the flats. Each block contains 210 axial holes. Fuel rods of compacted fissile and fertile particles are sealed into 132 of the holes, six holes are filled with a burnable poison (B_4C) to balance core neutronics and power distribution during operation, and the remaining 72 holes remain open as coolant (helium) channels. The 1160-MWe HTGR core will have an array of 3944 graphite-block assemblies in 493 vertical columns of eight assemblies each (Gulf Oil Corp. Facts).

It is planned (Gulf Oil Corp. Facts) that the fuel particles in the large (>1000 MWe), future HTGRs are to be a mixture of graphite-coated fertile and fissile BISO particles. An earlier report (DOCKET 50767-14) proposed that a silicon carbide coating (TRISO type) might act as an effective barrier to fission-product activities, including noble gases, and that the graphite in the assembly (and particles) might serve as a secondary barrier for metallic fission products. Also cited were experiments with multilayer pyrolytic-carbon-coated particles; these experiments indicated that the release fraction for fission products was the same as the failed-particle fraction, *i.e.*, about 0.8%.

2. Coolant Cycle

The helium primary coolant of a reference HTGR is contained almost entirely in the PCRV; a typical flow diagram of the coolant system is shown in Fig. 21. The two major coolant-system components are the helium circulators (compressors) and the helium-to-water steam generators. Cold helium leaves the steam generators and is forced (at $\sim 10^7$ lb/hr) through the annulus formed by the core barrel and the reactor-vessel wall and into the core-inlet plenum at the top of the core. The helium then flows down through the holes in the graphite assemblies and into the inlets of the steam generators at about 750°C and 700 psia (Gulf Oil Corp. Facts).

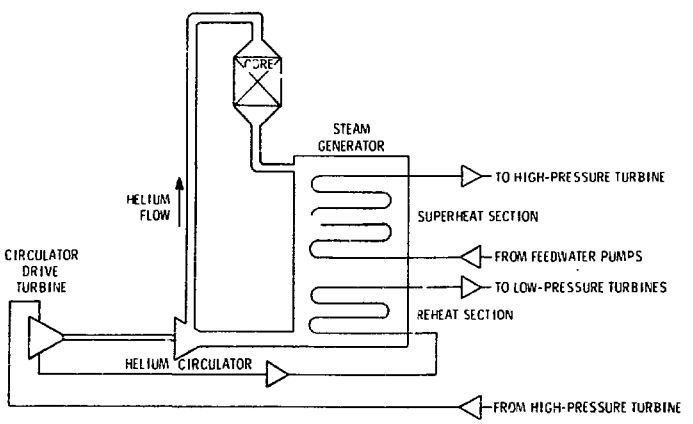

Fig. 21. Typical Flow Diagram for HTGR Plant (Gulf Oil Corp. Highlights)

An auxiliary to the primary coolant system is the helium purification system shown in Fig. 22. It is located in the head of the PCRV. The first stage is a high-temperature filter-adsorber which removes dust and volatilized metallic fission products from the helium side stream flowing at the rate of 8.4×10^4 liters (STP)/min in the reference plant (Gulf Oil Corp. Facts). The helium is next fed to a condenser that removes entrained water in the case of stream-generator tube failure. The cooled helium then flows through a dryer which removes any remaining water vapor and carbon dioxide. Leaving the dryer, the gas stream is chilled and fed to a low-temperature adsorber (activated charcoal) where any noble gases, carbon monoxide, nitrogen, or methane in the helium stream are removed. At this point, the helium purification stream first leaves the reactor vessel to be fed to a hydrogen getter unit (titanium-sponge getter at 370°C). Here, hydrogen and tritium are retained as a solid solution in titanium. The helium gas leaving the getter is highly purified and available for storage or recycle.

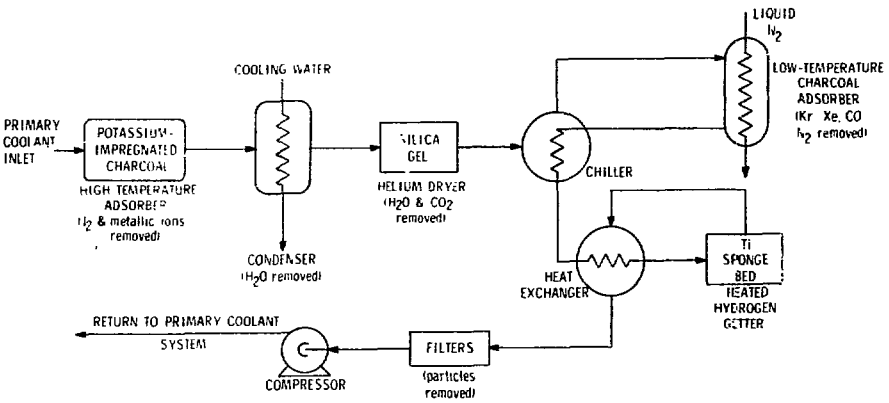

Fig. 22. Coolant Helium Purification System for HTGR Plants (Goodjohn)

Steam in a reference HTGR (Gulf Oil Corp. Facts) is produced in three-section steam generators (economizer-evaporator, superheater, and reheater sections) constructed of Incoloy and carbon and low-alloy steels. The steam-water cycle is similar to conventional turbine schemes using nuclear reheat. High-pressure steam (2500 psig at∿510°C) is supplied to the high-pressure turbine and reheat steam (574 psig at∿540°C) is supplied to the intermediate- and low-pressure turbines. High pressure turbine exhaust steam is used to drive the primary coolant helium circulators. The exhaust from the circulator turbines is then fed to the reheater sections of the steam generators.

3. Gaseous Effluent Systems

A gaseous waste-treatment system (Fig. 23) that would handle equipment and purge streams as well as gases produced by the regeneration of helium-purification adsorption beds is planned for reference HTGR plants (WASH-1085). The gases are piped to a common header, filtered, and monitored. If the radioactivity is such that release to the atmosphere is undesirable, the stream is

routed to a holding tank, compressed, and fed into a surge tank. The surge tank is sampled and the gas released at a metered rate through exhaust filters to the plant stack, utilizing the dilution by general ventilation air.

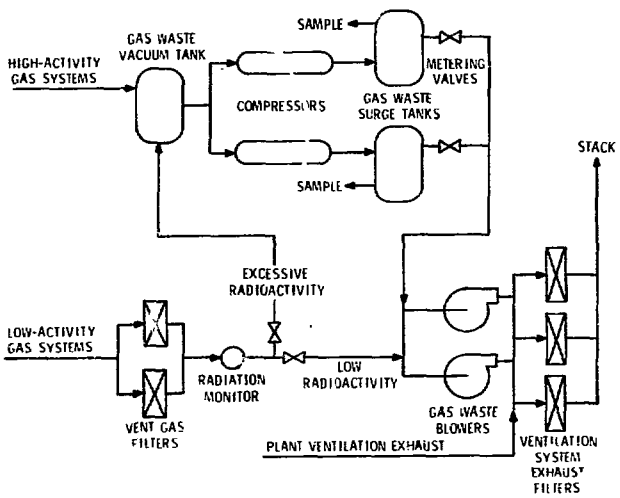

Fig. 23. HTGR Gaseous Waste-Treatment System (WASH-1085)

4. Liquid Effluent Systems

All aqueous wastes produced in the HTGR plant are to be collected and monitored for radioactivity. It is not expected (WASH-1085) that liquid wastes from the plant will contain any radioactivity, except during decontamination operations or possible accidents (such as steam generator tube rupture). Liquids that have radioactivity levels too high for discharge are to be decontaminated by a demineralization operation, monitored again, and discharged into a flowing-water canal.

B. Generation of Tritium in High-Temperature Gas-Cooled Reactors

The amounts of tritium (Table 23) that have been predicted from HTGR fuel and core components vary from ~1.4 x 10^4 Ci/1000 MWe-yr (Goodjohn, Kouts), from fission and from lithium and boron activation, to about 2.1 x 10^4 Ci/1000 MWe-yr, as calculated by two other sources (Colby, Snider). It should be noted that the latter amount is in agreement with the thermal neutron yield given for ^{235}U in LWR reactors (2 x 10^4 Ci/1000 MWe-yr). The amount of tritium that is produced solely as a result of fission has not been reported. A model used to calculate amounts of radioisotopes that could appear in HTGR fuel (Snider) includes neutron activation of assorted graphite impurities in addition to the fission process.

The $^3He(n,p)^3H$ reaction in the helium coolant of HTGRs is considered to be the major source of tritium (Gulf-GA-A12150). Estimates of the generation rate of tritium from the neutron activation of ^3He range from 3 x 10^3 (Goodjohn) to

6×10^3 Ci/1000 MWe-yr (Kouts). Another reference (DOCKET 50267-40) indicates that the water in the core-support floor and PCRV cooling systems will contain lithium hydroxide for pH control, resulting in probable tritium contamination of these systems due to the $^6Li(n,\alpha)^3H$ process (estimated at 9×10^{-3} Ci^3H/yr for the FSV HTGR).

TABLE 23. Estimated Generation Rate of Tritium and Appearance Rate in Helium Coolant of a HTGR

Source	Estimated Rate, Ci/1000 MWe-yr	
	Generation	Appearance in Helium
Fuel (fission and nuetron activation)	2.06×10^4[a]	1.6×10^2[b,d]
Coolant Helium (from neutron activation of ^3He)	3.3×10^3[c]	3.3×10^3[c]

[a]Calculated from Colby, Snider.

[b]Based on predicted 0.8% TRISO coating failure.

[c]Goodjohn.

[d]If based on recent test results (GULF-GA-A12222), this value might be as high as 6×10^3 Ci/1000 MWe-yr.

C. Appearance of Tritium in High-Temperature Gas-Cooled Reactor Waste Streams

1. Mechanisms and Amounts

Tritium is expected to appear in the helium-coolant waste stream of a HTGR. Investigations (Gulf-GA-A12150) of two mechanisms involving tritium transport in HTGR fuel have been made. The preliminary data (Gulf-GA-A12222) from experiments conducted to measure the amount of fission-product tritium that might diffuse through the coatings of fuel particles indicate that previous reliance on the imperviousness of the coatings may be unfounded. For reference fuel particles (Gulf Oil Corp. Facts), the new data show a tritium release of ~30% from both fissile-BISO particles containing $(Th,U)C_2$ and fertile BISO particles containing UO_2* at the temperature range of interest (900°C average fuel temperature in a projected 1160-MWe HTGR). It should be noted, however, that the percentages quoted were derived by comparing observed amounts of tritium remaining in the particles with calculated production rates. The fission yield used was 0.87×10^{-4} tritons per thermal fission of ^{235}U as predicted by Ray (Ray). More recent data (Fluss) shows a tritium yield of $2.9 (\pm 0.2) \times 10^{-4}$ tritons per ^{235}U fission with neutrons in the energy range from 170 to 700 keV. It is stated in a design description of a 1160-MWe HTGR (Wash-1085, Gulf Oil Corp. Facts) that from 15 to 20% of the neutrons appearing in the core flux

*Actual reference fertile fuel will be ThO_2 but no experimentation was cited for that material.

might have energies of 180 keV or more (8×10^{21} nvt). If the samples used in the experiments described above were irradiated in a neutron flux similar to that predicted for a large HTGR, then the tritium production rate might be twice that used in calculating release percentages. This would set the tritium release from tested fuel particles at ~65%.

Another potential mechanism for the appearance of tritium is being studied in an investigation (Gulf-GA-A12222) of the mechanism of hydrogen-tritium permeation through steam-generator materials. If permeation is observed, this mechanism would contribute to the tritium contamination of the steam-water system.

The problem of tritium diffusion through a HTGR vessel liner and the concrete of the PCRV may not yet have been considered, since no information on the subject appears to be available. Furthermore, there has not been an investigation of the tritium content of the methane that is said to be trapped out of the primary coolant by the helium-purification system (DOCKET 50267-15).

Based on the amounts of tritium formed in the helium coolant and that released from the fuel after failure, the primary coolant is estimated to contain 3-6 x 10^3 Ci/1000 MWe-yr. These figures appear to be low, however, in view of results that indicate there might be appreciable diffusion of tritium through the coatings of non-failed fuel particles.

An estimation of the amounts of tritium that might appear in the plant or be released to the atmosphere cannot be made until the result of work on diffusion of ^3H through reactor materials is reported. The systems for hydrogen-tritium removal (titanium getters) have been provided and will have the ability to remove H_2-HT from a stream containing up to 10 ppm of hydrogen (DOCKET 50267-44). Loaded getters are to be removed from the system for off-site retention, and hence, little tritium is expected to be released at the reactor to the atmosphere from the helium purification system.

To date, no HTGR of the size envisioned for the future has had operating experience. The 330-MWe FSV reactor, presently beginning operation, will provide the operating experience and opportunities for obtaining tritium transport data that may be useful for prediction of tritium behavior in 1000-MWe HTGRs.

An operating HTGR, the 40-MWe Peach Bottom No. 1 unit, is sufficiently different in core, vessel, and coolant loop design that it cannot be used as a model for the gas release expected from future HTGRs. For example, it is reported (Busch) that fuel failure in the reactor's first core was about 1.4%, almost twice that expected for hexagonal-block HTGR fuel elements (0.8%).

2. Potential Pathways

Fission-product tritium will permeate the graphite coatings of fuel particles, but the amounts have not yet been determined for conditions typical of a reference HTGR. It has been suggested (GULF-GA-A12222) that a portion of the escaping tritium will be adsorbed by the graphite matrix of the assembly. However, it is expected that some of the fission-product tritium will eventually appear in the helium coolant of the reactor. It is predicted that tritium will also appear in the helium coolant as a result of the $^3He(n,p)^3H$ reaction.

The possibility exists of diffusion of hydrogen and tritium through the ferrous metals of the steam generators, based on experience in other types of reactors. Present studies on the extent of this diffusion through HTGR materials (GULF-GA-A12150) are an indication of this concern. If this form of tritium transport does occur, then the pathways of the isotope through the steam-turbine system would be similar to those described for the BWR system.

Since the reheater sections of the steam generators produce steam at a pressure (∿600 psia) lower than the helium coolant pressure (∿700 psia), tube failure in these modules would result in leakage of contaminated primary coolant helium into the steam-water system (WASH-1085).

Other pathways to the environment could be conventional: leakage of the primary coolant helium and the steam-water systems. A schematic diagram of possible ^{3}H pathways is presented as Fig. 24.

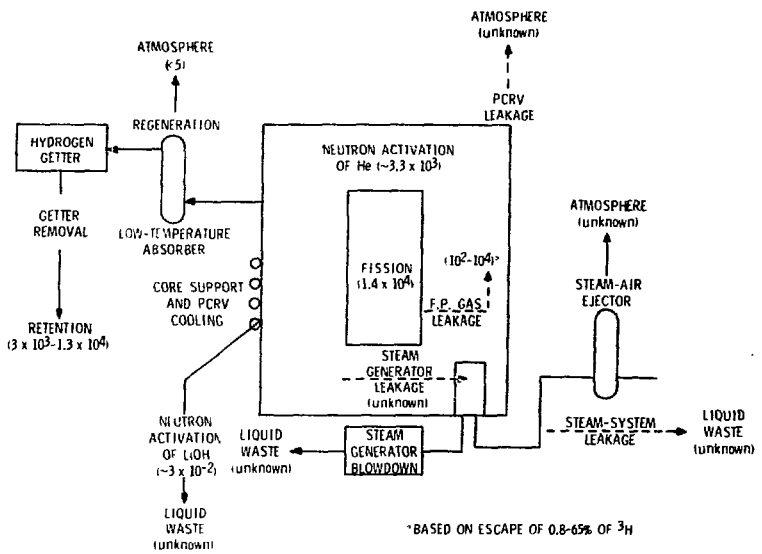

Fig. 24. Possible Paths of Tritium in a HTGR (amounts in Ci ^{3}H/1000 MWe-yr)

3. Effect of Management Schemes

The use of a helium purification system in the FSV HTGR (DOCKET 50267-44) and in advanced 1000-MWe HTGRs (Goodjohn) is a management provision that could have great effects on the release of tritium at HTGR plants. Helium purification is expected to reduce the equilibrium amount of tritium in the helium coolant by a factor of 10^{3}, from the predicted steady-state tritium content at FSV of about 6 x 10^{3} Ci without purification to about 6 Ci with the purification system in operation (DOCKET 50267-44). Most of the collected tritium will be bound chemically to the titanium sponge in the hydrogen-getter unit. The design description of the low-temperature-adsorption charcoal bed predicts that a six-month accumulation of tritium on the low-temperature charcoal bed, destined for the radioactive gaseous waste system (*i.e.*, plant stack) upon regeneration, would be ∿3 Ci (DOCKET 50267-44). The amount of tritium available for

annual release is that amount in the coolant (6 Ci) plus that deliberately liberated (6 Ci), or 12 Ci at FSV. The predicted amount of tritium release from a 1000-MWe HTGR (Goodjohn) is less than 5 Ci/yr. The balance of the tritium, then, is collected by the titanium getter ($<10^3$Ci/yr for FSV, 3.3×10^3 Ci/yr for a reference 1000-MWe HTGR).

4. Conclusions

If the helium purification system designed for Fort St. Vrain and future HTGRs performs as designed, the appearance of tritium outside the reactor vessel (except for that disposed of as titanium tritide) may be <12 Ci/year for a 1000-MWe HTGR. An unpredicted increase of tritium in the primary coolant could be managed by upgrading the hydrogen-removal unit of the purification system. From indications in the design reports, the low partial pressures of hydrogen in the coolant would probably preclude any occurrence (except for reheater-tube failure) of tritium in the steam-water system and, hence, in the liquid effluents where tritium is commonly found in LWRs.

D. Generation of Noble Gases in High Temperature Gas Cooled Reactors

Radioactive noble gases will be formed in the HTGR only by the fission process. Hence, the appearance of these gases in HTGR effluents is dependent on the extent of fuel-coating failure. Based on computer calculations (Snider), the amount of ^{85}Kr that might be generated in HTGR fuel with a burnup of 90,000 MWd/metric ton would be about 1×10^4 Ci/metric ton or 3.4×10^5 Ci/1000 MWe-yr. Assuming a fuel-failure factor of 0.008, the amount of ^{85}Kr in the coolant of a 1000-MWe HTGR should be about 2.7×10^3 Ci/yr. Estimates of ^{85}Kr release from the Fort St. Vrain plant are confusing. The Environmental Statement (DOCKET 50267-40) fixes the anticipated release from the regeneration of the low-temperature adsorbers* of the helium purification system at about 960 Ci/yr. The safety analysis report (DOCKET 50267-44) states that the expected amount is 560 Ci/yr.** These differences will no doubt be resolved by measurement during actual operation of the plant.

E. Appearance of Noble Gases in High Temperature Gas Cooled Reactor Waste Streams

1. Mechanisms and Amounts

Noble gas fission products are dispersed in the helium coolant and collected in the helium purification system's low-temperature charcoal adsorber. Two other transport routes are cited, however (DOCKET 50267-40). Leakage through the reactor vessel PCRV of the FSV 330-MWe plant is predicted to be at a rate of 28 Ci of noble gas per year. Another possibility arises when tube failure occurs in the reheater section of the steam generator. Reheat steam is at lower pressure than is the helium coolant. Noble gas release due to this mechanism is expected to be about 5 Ci/yr through the plant's condenser air ejector and feedwater deaerator. A similar occurrence is expected as a result of failure of water piping that cools the core support floor and PCRV walls. Also mentioned are various cask, fuel-handling-component, and waste-tank-vent purges (DOCKET 50267-40).

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*Assumed 99% efficient, thus trapping all the ^{85}Kr released into the coolant.

**Design activity is stated to be 4.8×10^3 Ci ^{85}Kr/yr.

Radioactive noble gases appear primarily in the low-temperature adsorber in the helium purification system. Noble gases are collected in the adsorber during six months of operation and isolated for a minimum of 60 days (for decay of short-lived noble-gas species); the adsorber is then regenerated using a combined heatup and helium purge.

The environmental statement for the FSV HTGR (DOCKET 50267-40) presents amounts of radioactive noble gases that are anticipated to be released at various sites within the plant. These data are presented in Table 24. The total amount (from Table 24) is slightly more than 990 Ci per year, the bulk of which (∿950 Ci/yr) is ^{85}Kr. Another recent report (DOCKET 50267-44) states that from the regeneration of the low-temperature adsorbers alone, the plant might release an expected activity of 560 Ci/yr (∿99% ^{85}Kr).

TABLE 24. Anticipated Annual Releases of Fission-Product Noble Gases in Effluents from the 330-MWe Fort St. Vrain Reactor (DOCKET 50267-40)

	Noble-Gas Release, Ci/yr			
Isotope	From Reactor Building Leak	From HPS[a] Regeneration	From Air Ejector	Total
^{85}Kr	0.09	947[b]	0.002	947
^{87}Kr	2.0	-	1.2	3.2
^{88}Kr	11.0	-	3.2	14.2
^{131m}Xe	0.04	4.8[b]	0.002	4.8
^{133}Xe	12.0	9.0[b]	0.6	21.6
^{135}Xe	2.4	-	0.2	2.6
^{138}Xe	0.01	-	0.03	0.04
Totals	27.5	960.8[b]	5.2	993.4

[a]Helium purification system low-temperature adsorber.

[b]After 60-day delay period.

There has been no experience with the FSV and 1000-MWe HTGRs. If actual fuel failure experience (1.4%, see Section V.C.2) at the Peach Bottom 1 HTGR (Busch) is used, along with computer predictions of noble gas generation in typical HTGR fuel (Snider), then ^{85}Kr generation of 3.4×10^5 Ci/1000 MWe-yr would give a ^{85}Kr appearance rate in the coolant of a reference 1000-MWe HTGR of 4.7×10^3 Ci/yr. This number is very close to the stated design limit (4.8×10^3 Ci/yr) given for the 330-MWe FSV HTGR (DOCKET 50267-44).

2. Potential Pathways

Since noble gases are produced solely within the fuel and are assumed to be contained there by fuel coatings, the only pathway for noble gases to the helium coolant is through coating failure. Unlike the case of tritium, no diffusion of noble gas through vessel and component materials is expected. Noble

gases can, however, appear in the water-steam system via defects in tubes of the reheater modules of the steam generators. Again, through the defect route, noble gases could also appear in the water stream of the reactor structural cooling system (PCRV and core support floor). The regeneration of the low-temperature adsorbers will release the greatest quantity of noble gases, however, and, owing to decay, the bulk will be long-lived ^{85}Kr.

3. Effect of Management Schemes

Delay (60-day minimum) of the release of adsorbed gases in the low-temperature adsorbers of the helium-purification system prior to regeneration discharge will ensure the decay of all noble gases except ^{131}Xe, ^{133}Xe, and ^{85}Kr (DOCKET 50267-44). Of these gases, ^{85}Kr contributes over 99% of the total activity. It has been suggested (Goodjohn) that this stream could be compressed and pumped into standard gas cylinders for off-site retention. Goodjohn also states that additional equipment can be provided for the helium-purification system to return all of the ^{85}Kr trapped in the low-temperature adsorbers to the primary coolant stream. In this case, the adsorbers would be sized to hold all the ^{85}Kr that might be released to the helium during the life (30 yr) of the HTGR plant.

4. Conclusions

Conclusions about the appearance of noble gases in a HTGR plant will have to wait until performance data are made available from tests or operation. Based on Gulf General Atomic calculations, however, the proposed use of a helium-purification system would keep the amounts of noble gas activity in the primary coolant to one-third of that present if the purification system were off stream. As seen in Table 25 (DOCKET 50267-44), the ^{133m}Xe would be reduced by a factor of 13, ^{133}Xe by 30, ^{131m}Xe by 60, and ^{85}Kr by nearly 1000.

Krypton-85 is the major noble gas released to the atmosphere during regeneration of the low-temperature adsorbers. This release can be avoided either by storing the gas in standard gas cylinders or by returning it to the primary coolant stream.

The appearance of short-lived noble gases outside the PCRV would be the result of primary coolant leakage through defects in the vessel, steam generators, compressor bearings, or core support floor and PCRV cooling piping. Release of the short-lived isotopes through these streams could be minimized, however, by tank hold-up and sampling, followed by controlled release into the reactor building vent or stack.

TABLE 25. Equilibrium Amounts of Noble Gases in Coolant of the Fort St. Vrain Reactor (DOCKET 50267-44)

Isotope	Half-life	Equilibrium Amount, Ci	
		Without Helium Purification System	With Helium Purification System
^{83m}Kr	1.86 h	2380	1710
^{85m}Kr	4.48 h	3860	1990
^{85}Kr	10.76 yr	3490	3.84
^{87}Kr	76 m	3440	2690
^{88}Kr	2.8 h	6420	4020
^{89}Kr	3.16 m	1200	1190
^{90}Kr	32.3 s	710	709
^{91}Kr	9 s	375	375
^{131m}Xe	11.99 d	157	2.51
^{133m}Xe	2.23 d	368	28.8
^{133}Xe	5.29 d	19800	695
^{135m}Xe	15.3 m	1110	1050
^{135}Xe	9.17 h	4280	1430
^{137}Xe	3.84 m	666	657
^{138}Xe	14.2 m	871	822
^{139}Xe	39.7 s	465	464
^{140}Xe	13.6 s	298	297
Total		49890	18134

APPENDIX

Physical and Radiochemical Information Pertinent To The Consideration of Tritium and Noble Gas Management

TABLE A-1. Physical Properties of Relevant Atmospheric Gases

Property	Kr	Xe	Ar	O_2	N_2
mp[a] at 1 atm (°C)	-157.1	-111.8	-189.2	-218.4	-209.9
bp[a] (°C)	-152.9	-108	-185.9	-183	-195.8
Critical Temp.[a] (°C)	-63.8	+16.6	-122.5	-118.4	-147.2
Critical Press[a] (atm)	54.3	58	48	50.1	33.5
Heat of Vaporization at bp[a] (cal/mol)	2310	3110	1554	816	1336
Density of Liquid at bp[b] (g/cm^3)	2.41	3.06	1.39	1.14	0.81

[a]Matheson

[b]Calculated from data in CGA.

TABLE A-2. Properties of Isotopic Hydrogen[a]

Property	H_2	D_2	T_2	HD	HT	DT
bp[b] (°K)	20.39	23.67	25.04	22.14	22.92	24.38
Triple Point[b] (°K)	13.96	18.73	20.62	16.60	17.62	19.71
Pressure at Triple Point[b] (mm Hg)	54.0	128.6	162.0	92.8	109.5	145.7
Critical Temp.[b] (°K)	33.24	38.35	40.44	35.91	37.13	39.42
Critical Pressure[b] (mm Hg)	9736	12487	13878	11134	11780	13300

[a]Mittlehauser

[b]Values quoted here are for the normal state, *i.e.*, for the high-temperature ortho-para composition.

TABLE A-3. Physical Properties of Isotopic Water

Property	H_2O	D_2O	T_2O	HDO	HTO	DTO
bp (°C)	100.00[a]	101.42[a]	101.51[a]	-	100.8[b]	-
Vapor Pressure at 25°C (mm Hg)	23.76[b]	20.6[b]	19.8[b]	22.1[c]	21.7[b]	20.2[c]
Triple Point (°C)	0.010[a]	3.82[a]	4.49[a]	2.15[c]	2.67[c]	4.19[c]
Pressure at Triple Point (mm Hg)	4.58[a]	5.02[a]	4.92[a]	-	-	-

[a]Jones.

[b]Dirian.

[c]Calculated from work of Van Hook.

TABLE A-4. Radioactive Properties of Tritium and ^{85}Kr

Property	^3H	^{85}Kr
Half-Life (yr)	12.262[a], 12.35[b]	10.76[a]
Decay characteristics[a]	β, 18.6 keV max., 5.7 keV avg.	β, 0.67 MeV max., 0.249 MeV avg. γ, 0.514 (0.41%)
Specific Activity[c]		
Ci/g	9700	390
Ci/cm^3(STP)	2.6 (T_2, T_2O) 1.3 (THO, DTO)	1.5
Heating Effect[c]		
W/g	0.32	0.56
W/liter(STP)	0.085(T_2, T_2O) 0.044 (THO,DTO)	2.1

[a]Lederer.

[b]Moghissi.

[c]Calculated from data in Lederer.

TABLE A-5. Maximum Permissible Biospheric Concentrations of Tritium and ^{85}Kr[a]

Media	μCi/ml	
	^{3}H	^{85}Kr
In air	2×10^{-7}[b]	3×10^{-7}[c]
In water	3×10^{-3}[b]	-

[a]From Ref. 10CFR20, Appendix B, Table II, 168 hr/week exposure.

[b]For soluble species.

[c]For submersion in a hemispherical infinite cloud.

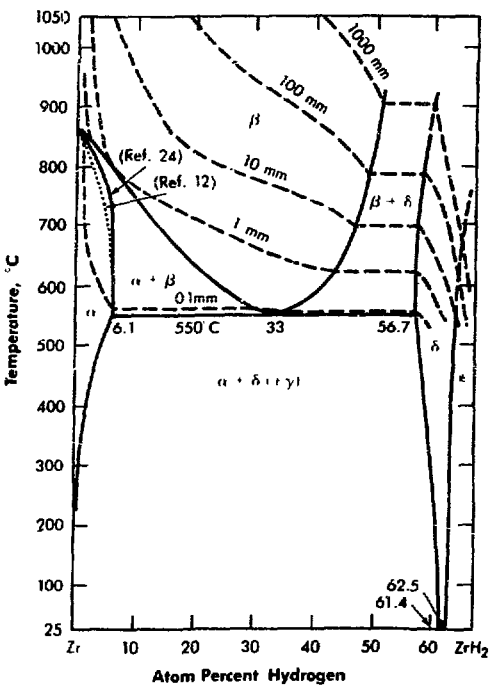

Fig. A-1. Zirconium-Hydrogen Phase Diagram (Mueller).

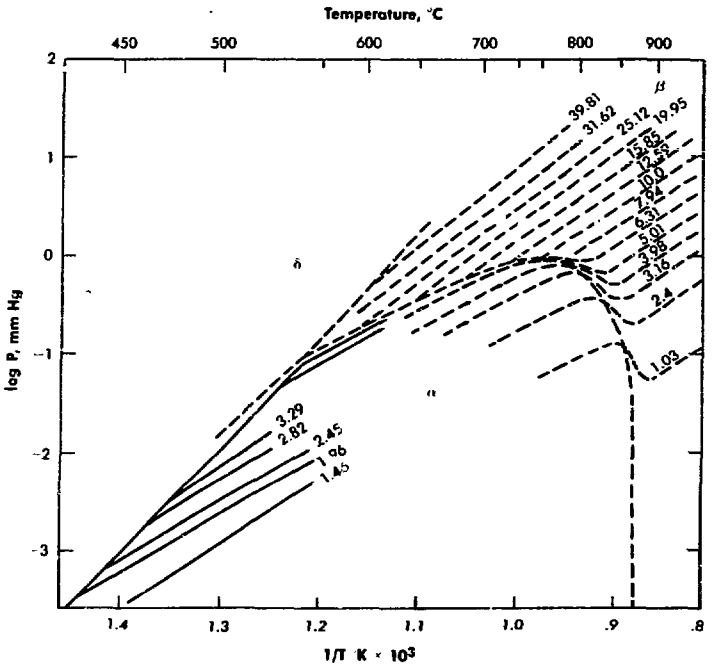

Fig. A-2. Pressure-Temperature Isochores for Zirconium-Hydrogen System (Mueller).

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